Extreme sensitivity of plasmon drag to surface modification

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Abstract Plasmon-induced photocurrents in 1D profile modulated structures switch their polarity in the presence of an additional monolayer at the metal-dielectric interface. The effect presents opportunities for compact plasmonic sensors with electrical detection.

OCIS codes: (280.4788) Optical sensing and sensors; (260.3910) Metal optics; (240.6680) Surface plasmons; (130.6010) Sensors

Plasmon drag effect (PLDE) is a giant enhancement of photocurrents under plasmon resonance conditions [1-2]. It provides an opportunity to monitor plasmonic excitations electrically, and is promising for plasmonic based sensing with compact electric detection [3]. According to predictions of electromagnetic momentum loss approach, the effect may be sensitive to surface modification, since the effective "plasmonic pressure" force acting on electrons is confined to the surface layer [4]. In the experimental work [5], photocurrents in plasmonic films strongly decrease in magnitude and switch their polarity when the air is pumped off. This points to the significant role of surface charges in the generation of photoinduced voltages.

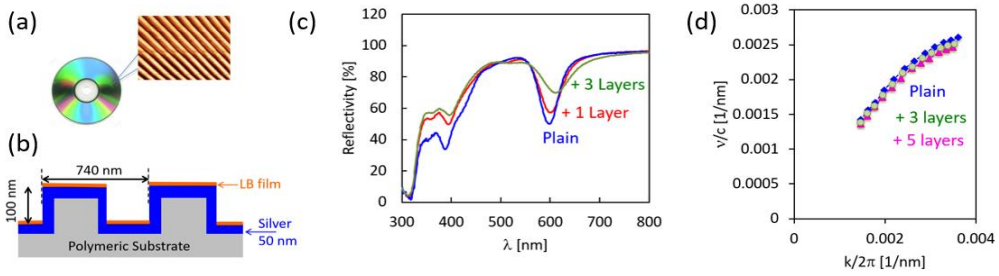

Fig. 1. (a) Nanostructured substrate (AFM image) is derived from Verbatim DVD-R; (b) Schematic of the sample with mono- or multilayer film deposited with LB method; (c) Reflectivity spectra at p-polarization before (plain) and after deposition of LB films; (d) Dispersion curves for 1-d order SPP with different number of layers

In order to better understand the role of metal dielectric interface in PLDE, we study photocurrents in plasmonic structures without and with one or several monolayers deposited onto the surface. As a convenient system for our experiments, we choose a silver surface with 1D modulated profile, where surface plasmon polaritons (SPPs) can be excited by direct illumination at the coupling conditions, $k_{SPP} = k_0 \sin\theta + m*2\pi/d$. Here k_0 is the optical k-vector, θ is incidence angle, $m \neq 0$ is an integer, d is grating period; $k_{SPP} = k_0\sqrt{\frac{\varepsilon_m\varepsilon_d}{\varepsilon_m+\varepsilon_d}}$ is the SPP k-vector, ε_m and ε_d are the permittivity of the metal and dielectric, respectively [6]. The experimental sample (see Figs. 1 (a) and (b) for the schematics) consists of a polycarbonate substrate (derived from a commercial DVD) having concentric periodic grooves with the periodicity d = 740 nm and modulation height of ~ 100 nm, and a thin film of silver (~50 nm) deposited on the top. Monolayers of stearic acid are deposited onto the silver surface using Langmuir-Blodgett (LB) technique (via top-down deposition). The LB technique allows one to fabricate films of closely packed molecules with well-controlled thickness, and stearic acid (SA) is one of the most commonly and widely used materials for this deposition method [7]. However, since our structures are strongly modulated, it is not clear how well the monolayer film follows the profile. One can assume that it covers flat (horizontal) areas of the grating (Fig 1 (b)).

Fig 1 (c) shows the reflection spectra recorded at p-polarization in the sample before and after addition of SA monolayer/s. The SPP (first order, m = 1) is seen as a pronounced dip in the reflection at ~ 600 nm at 15 deg of incidence. The addition of monolayers broadens the resonance and shifts its spectral position to higher wavelengths. Fig. 1(d) shows the SPP dispersion curves estimated from the angular dependence of the dip in the samples with various number of layers. The curves are typical for SPP and are close to each other for the samples with 0-5 layers.

For PLDE experiments (Fig. 2 (a)), the sample is prepared as a strip of 3 mm thick and 15 mm long with vertical orientation of the grooves. The electrical contacts are placed at the opposite ends. The sample is placed on the

goniometer stage and illuminated with the p-polarized pulsed laser light at the wavelength λ = 532 nm, pulse duration of 10 ns and energy of ~0.3 mJ per pulse. The photoinduced electric voltages are recorded using Tektronix 3 GHz oscilloscope with 50 Ω internal resistance. In the experiments, first, the PLDE voltage U is measured in the sample before the deposition of a monolayer (plain) for various angles of incidence, θ. Then the measurements are repeated in the same structure after each deposition of SA layers. Thus, all the data in Figs. 2 (b, c) are obtained from the same sample. This allows us to exclude other factors such as variations in the sample geometry, which can affect its electric behavior.

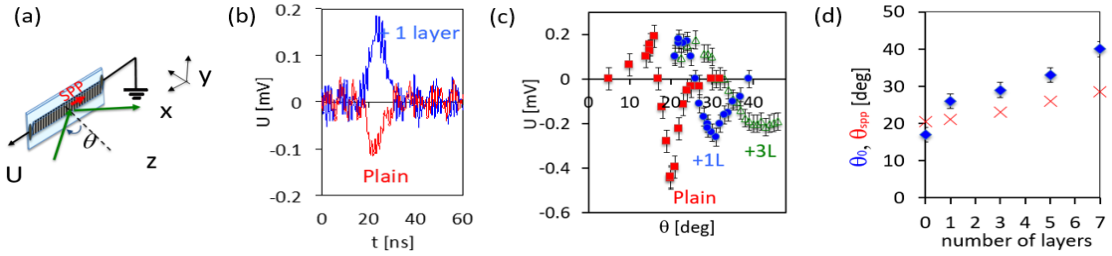

Fig. 2. PLDE in DVD + LB structure: (a) Experimental setup; (b) PLDE signal at 23° before and after deposition of SA monolayer; (c) Magnitude of the photoinduced voltage vs incidence angle before (plain) and after deposition of 1 (+1 L) and 3 (+3 L) SA layers; (d) Dependence of the switching angle (blue) and SPP resonance angle (red) on the number of monolayers

While the addition of a single SA layer does not significantly change the optical properties (Figs. 1 (c, d)), it strongly affects photoinduced voltages. As shown in Fig 2 (b), adding a monolayer switches the polarity of the PLDE signal to the opposite polarity. Before the deposition (in the plain sample), electrons are dragged in the direction of the k-vector of the incident photon while in the sample with a monolayer, electrons move against the k-vector. Such an ultimate change in the PLDE signal is observed at a certain range of the angles of incidence θ = ~ 20° - 25°. It can be explained by taking into account the following experimental findings: (i) $U(\theta)$ in our structures has a strong asymmetric shape, demonstrating switching in the signal polarity at a particular angle, θ_0; (ii) The angle θ_0 shifts toward higher angles upon the deposition of LB layers (see Fig. 2 (c)).

Such an asymmetric Fano-like resonance shape of the angular dependence of photo-voltages is observed in the systems with strongly modulated profiles in [8], where the polarity-switching phenomenon is tentatively ascribed to coupling of SPPs and localized plasmon resonances. According to the data presented in Fig 2 (d), θ_0 is close to the SPP resonance angle, θ_{SPP}, but grows faster than that with the increase of the number of layers. This discrepancy can be associated with various factors, including changes in the effective roughness of the surface. Theoretical analysis is in progress.

In conclusion, the PLDE signal in plasmonic gratings is very sensitive to the surface modification. Excited under a certain angle, the photocurrents switch to the opposite polarity upon the addition of a single monolayer onto the surface. The effect presents interest for applications in nanoscale sensing.

This work is supported by National Science Foundation (NSF) (1830886), Air Force of Scientific Research (AFOSR) (FA9550-18-1-0417) and Department of Defense (DoD) (W911NF1810472).

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