Plasmon drag effect in metal nanostructures

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**Abstract.** In order to better understand the mechanism of the photon drag effect in plasmonic nanostructures, photo-induced electric signals have been studied in gold and silver films and various plasmonic nanostructures. The spectral dependence of the effect points to the primary role of individual localized plasmon resonances in the photo-induced electromotive force (emf) generation responsible for the photon drag effect. We demonstrate the potential to engineer both the magnitude and polarity of the emf with nanoscale geometry and provide a simple model based on the intrinsic nonlinearity of metal in defining this effect.

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1. Introduction

Recent advances in the field of plasmonics have continued the expansion of the great promises for future electronics, allowing one to combine the inherent high operational frequency associated with optical wavelengths with nanoscale device dimensions, something that is not possible with standard optics due to the limits of the diffraction condition [1, 2]. Plasmonic components of different functionality have been proposed [3–6]. In order to incorporate such elements into electronic circuitry, and to control or monitor them electrically, the intricate direct coupling of photons, electrons and surface plasmons is of crucial importance. The photon drag effect (PDE), i.e. generation of electric currents in response to light illumination, is one manifestation of such coupling. In the classical approximation, the photon drag is often discussed in terms of a momentum transfer from incident photons to free conduction electrons (light pressure) [7]. The efficiency of this classical process is quite low due to the large disparity in masses of interacting particles. In semiconductors where photon drag is significant, other mechanisms enabling this effect were shown to play a major role, such as momentum-selective interband or intraband transitions and different mobilities in corresponding bands [8–11].

In bulk metals, the PDE is very small [12, 13]. However, it was theoretically predicted that the PDE would be significantly enhanced in metal nanowires due to the effects of the high intensity and high gradients of light intensity, associated with plasmon propagation in confined geometry [14]. Experimentally, the influence of surface plasmons upon photo-induced currents was first studied in thin metal films in [15, 16] using the Krechmann geometry [17], which is commonly used for launching surface plasmon polaritons (SPP) [18] on metal surfaces. Photo-induced currents observed at the SPP resonance condition exceeded the theoretical estimations predicted by the light pressure mechanism by over an order of magnitude, even when taking into account the increased absorption that occurs at plasmon resonance conditions, thus demonstrating the potential for the PDE in nanophotonic applications [16]. On resonance, electron drag was observed to flow in the direction of the optical k-vector projection on the film surface, \( k_x \). However, a smaller signal of the opposite polarity (electron drag in the \( -k_x \) direction) was observed at illumination off the resonance condition. This off-resonance signal was tentatively ascribed to SPPs as well, which might be excited in the \( -k_x \) direction due to the surface roughness [16]. Photon drag in thin metal films was also studied in the conditions of spatially modulated illumination [19] and in surfaces with an asymmetric profile [20]. These
studies clearly showed that the strong PDE observed in thin metal films was primarily related to the excitation and propagation of SPPs. In [19, 20], electrons were dragged in the direction of the SPP propagation, regardless of the direction of the light pressure force originated from photons, thus further demonstrating that the effect is dominated by plasmon propagation.

In order to better understand the mechanism of the plasmon-related optical drag (PLDE), and to explore the possibility that this effect can be directly controlled through the engineering design of the nanoscale geometry, here we report on the PLDE in several nanostructured systems as a function of (i) surface roughness and topology, (ii) angle of incidence and (iii) wavelength of illumination.

2. Experimental

Three experimental systems were explored within this investigation and are described as follows:

1. 2 mm × 10–12 mm strips of flat, 30–60 nm thick gold and silver films. The films were deposited on glass substrates via thermal evaporation. The experimental samples include both as-deposited films along with those featuring random surface roughness. In order to produce this roughness, the as-deposited films were exposed to intense laser light of wavelengths between 530–550 nm and intensities up to 20 mJ cm$^{-2}$. The level of exposure was chosen in such a way that it did not produce any significant visible damage, but resulted in the creation of nanoscale holes, as shown in the SEM image in figure 1(a). As an alternative method to create roughness, gold nanoparticles (5–50 nm in size) were randomly dispersed on the surface of a gold film.

2. Similar sized strips of gold and silver quasi-periodic nanomeshes (or fishnets). These samples were produced by thermal deposition of metal onto porous anodic-alumina membranes (AAO) of different periodicities (30–250 nm), as shown in the SEM image...
Figure 2. (a), (b) SEM of the arrays with the periods of 250 and 500 nm; (c) schematic of the sample.

in figure 1(b) with a schematic of the deposition process provided in figure 1(c). In order to introduce an asymmetry at the nanoscale, some samples were fabricated using oblique-angle metal deposition.

3. Periodic arrays of gold-coated silicon nanopillars on silicon substrates. Si nanopillars, 60 nm tall and 75 nm in diameter were situated in a square array with a periodicity of 250 or 500 nm (referred to as Array 1 and Array 2, with corresponding SEM images presented in figures 2(a) and (b), respectively). The Si nanopillars were fabricated via the electron beam lithography and reactive ion etching. A full description of the fabrication process is provided in [21]. The nanopillar arrays were 0.5 mm on a side. A stencil mask was created to enable the continuous coating of the Si nanopillars with a 50 nm thick Au film via e-beam evaporation that extended only to the edges of the arrays in one direction, but provided electrical leads to the ends of the sample, as shown in figure 2(c).

The experimental setup for measuring the PLDE effect is shown in figure 3. The sample was placed on a rotating stage and illuminated with p-polarized laser light (∼5 ns pulses of an optical parametric oscillator) at different angles of incidence. The wavelength of illumination, λ, was varied in the range between 420 and 750 nm using the output of an optical parametric oscillator (OPO) tunable laser system. In order to perform a measurement at shorter wavelengths, we used the third harmonics of a Nd:YAG laser with λ = 355 nm and approximately the same pulse duration. The diameter of the excitation spot was about 2.5 mm, which is slightly larger than the width of the metal strip and larger than the size of a nanopillar arrays. Two electric contacts were attached using the conductive paste to the opposite sides of the sample, and electrically connected to Tektronix 3 GHz oscilloscope with 50 Ω input impedance. The pulsed illumination induced an electric signal with its temporal profile approximately corresponding to that of the laser pulse.
3. Results

3.1. Effect of surface roughness

The optically induced electric signals were relatively weak in nominally flat gold films and were dramatically enhanced in rough and nanostructured samples. As a rule, the magnitude of the electrical signals grew approximately linearly with an increase in laser power, saturating at high illumination intensities close to the film damage threshold. In figure 4(a), the magnitude of the photo-induced voltage was recorded as a function of the laser pulse power, both in an as-deposited nominally flat gold film and, then, in the same film after 5–50 nm gold nanoparticles were randomly deposited on the film surface. As a result, the presence of the Au nanospheres induced an approximately two-fold increase of the slope of the power dependence. Further dramatic increases in the signal magnitude were observed in the gold nanomesh strips, as shown in see figure 4(b), where the data obtained in the gold nanomesh is shown together with the data from the flat films of figure 4(a). Practically no signal was observed in a flat silver film as fabricated. However, films with laser-induced roughness demonstrated photo-induced voltages of a significant magnitude, figure 4(c).

In an effort to clearly distinguish the role of the nanostructure in providing the amplified effect, the photo-induced voltage was measured as a function of laser spot position on the Au-coated silicon nanopillar arrays. Presented in figure 5 is the photo-induced voltage response to the laser illumination recorded at three different spatial positions of the laser illumination spot; with the laser spot focused directly on the array (green curve), on the flat gold film serving as the left electrical contacts (purple curve) and the gold film to the right (blue curve) of the
array. When the nanopillar array pattern was illuminated, the signal was drastically enhanced in comparison to the surrounding flat gold films and was much higher than signals observed in the other nanostructured samples under similar experimental conditions. Thus, rough and nanostructured surfaces clearly not only exhibit the PLDE effect, but provide a mechanism for the enhancement of this photo-induced voltage in comparison with that observed at flat metal surfaces.

3.2. Angular dependence of PLDE

Typical dependences of the photo-induced voltage on the angle of incidence, $\Theta$, in rough, but nominally flat films are shown in figures 6(a) and (b). The dependences can be approximated with the sinusoidal function of $\Theta$, reaching a maximum near 90°. The signals in the Au-coated Si nanopillar arrays were observed to grow with increasing angle, reaching a maximum near $\sim$50°, and sharply decreased with the further increase in the incidence angle (figure 6(c)). Note that in contrast to the case of the metal films, the total light energy incident on the array was decreased upon increase of the angle due to a restricted size of the array (0.5 mm), which was
Figure 7. The PLDE magnitude versus angle of incidence in Au on AAO deposited at (a) normal deposition angle, periodicity of AAO of $d = 250$ nm and oblique deposition angles of (b) $\sim 20^\circ$ ($d = 250$ nm) and (c) $\sim 30^\circ$ ($d = 150$ nm). The wavelength of illumination is indicated.

smaller than the diameter of the laser spot. This can contribute to, but not fully explain the sharp drop of the signal at higher angles.

As a rule, in flat films, the polarity of the photo-induced current corresponded to the electron drag in the direction of $k_x$. In some rare cases (in particular, when a freshly made film was illuminated from underneath, through the glass substrate), we observed a small signal in the opposite direction, anti-parallel to the $k$-vector projection. In rough films, the electrons were always dragged in the direction of $k_x$. Therefore, not surprisingly, there was no signal at the normal incidence. However, this was not the case for nanomesh samples. While the nanomesh fabricated at the normal deposition angle exhibited similar behavior to the flat films, with the electron drag being parallel to the $k$-vector projection, there was a slight asymmetry in the angular dependence and a small non-zero signal at the normal incidence (see figure 7(a)). Similar to the results of [20], such a dependence can be due to a slightly asymmetric profile of the sample and excitation of plasmons propagating predominately in one direction. In order to test this assumption, we fabricated strips with asymmetric profiles, setting an AAO substrate under an oblique angle during metal deposition.

As shown in figures 7(b) and (c), unidirectional PLDE was observed in the samples fabricated with oblique deposition. In that case, the electron drag was only observed in one direction regardless of the direction of the optical $k$-vector, and a substantial photo-induced voltage was observed even at normal incidence.

3.3. Spectral dependence of PLDE

Figure 8 shows the spectral dependence of PLDE in various samples, including a rough silver film (with the laser-induced nanoscale surface damage), quasi-periodic gold and silver nanomesh samples, and the two Au-coated Si nanopillar arrays. The amplitude of the signal is normalized to the pulse energy incident on the sample. As one can see, the spectral dependence is non-monotonous, with a maximum in the range of $\sim 440$ nm in silver samples, $\sim 550$ nm in gold nanomesh and around 610 nm in nanopillar samples. In all the cases, the peak of PLDE was in the range of the localized plasmon resonance as measured via transmission or reflection spectra. The characteristic signatures of the plasmon resonance in silver and gold nanomeshes were observed in the optical transmission spectra shown in the corresponding figures. The reflection spectra of the gold nanopillar array had several features due to its periodic structure.
Figure 8. Spectral dependence of PLDE in (a) silver nanomesh with periodicity of 30 nm (Ag1), and a roughened silver film (Ag2); (b) gold on AAO with periodicity of ∼250 nm (Au1) and 150 nm (Au2); (c) nanopillar Array 1 (A1) and Array 2 (A2). Solid traces are the corresponding transmission (T) and reflection (R) optical spectra.

The feature around 600 nm corresponds to the localized plasmon resonance of an individual gold nanopillar [21, 22]. Note, that in the nanopillar arrays, the magnitude of PLDE is almost twice as large in the array with the 250 nm pitch in comparison to the 500 nm pitch, which is roughly consistent with the increase in the filling fraction of nanopillars within these arrays.

4. Discussion

As was discussed earlier [16, 19], the effect observed was orders of magnitude stronger than was predicted using a simple light pressure model, [7] even when taking into account the increased absorption due to the plasmonic resonance conditions. As was found in our experiments, the spectral dependence of the photo-induced voltage reached a maximum at an incident wavelength close to the localized surface plasmon resonance of the individual nanostructures, indicating the predominant role of the nanoscale features. In addition, the nanopillar array with the smaller pitch provided approximately twice the magnitude of the photo-induced voltage in comparison to the nanopillar array with the larger periodicity. This difference in pitch corresponds to roughly twice the number of nanopillars along the current path, again clearly illustrating the role that the individual plasmonic resonators play in governing this effect. These observations taken together can be generally described with the following scenario: each nanopillar when excited with light, works as a source of the electromotive force (emf) (see the illustration in figure 9).

Using the data from figure 5, and taking into account the total size of the array, (500 µm), the emf of an individual nanopillar was in the microvolt range for a light intensity of 2 MW cm$^{-2}$. 

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Figure 9. (a) Equivalent circuit of PLDE in nanopillar arrays. (b) Asymmetry of positive and negative charges motion under driving field in nanostructure according to Panasyuk et al [25]. (c) SEM image of the gold nanomesh on AAO fabricated under oblique deposition. Arrow shows the direction of the photoinduced electron drag.

We believe this effect is associated with intrinsic nonlinearity of nanostructured metal. As it was discussed in [23, 24] for nanostructured samples, sharp boundaries can lead to second harmonic generation with the efficiency proportional to the surface to volume ratio. The nonlinearity of metal nanoparticles was also discussed in [25] where electrons were considered within the framework of the Drude model as a negatively charged compressive fluid. Following [25], under the periodic field, \( E_0 \cos(\omega t) \), negative charges can be compressed indefinitely on one side of the particle, while a positively charged layer with the width, \( h \), is formed on the other side. In a slab geometry (figure 9(b)), the width of the positive layer, \( h \), can be estimated as \( h = |\sigma|/\rho \), where \( \sigma = \varepsilon_0 \Lambda(\omega) E_0 \cos(\omega t + \phi) \) is the charge induced by the optical field, \( \rho = Ze/l^3 \) is the background positive charge density, \( Z \) is the number of electrons per atom, \( e \) is the electron charge, \( l \) is the lattice constant, \( \phi \) is the phase shift and \( \Lambda(\omega) \) is the resonant factor. As a result, the range of the oscillations of the positive charge is different from that of the negative charge. According to Panasyuk et al [25], such a difference can result in the generation of high harmonics.

Let us now suppose that motion of the charges in a nanoparticle under the driving field is highly asymmetric. Asymmetric boundary conditions can be one of possible sources for such asymmetry. As an approximation, let us assume that a positively charged layer with the width \( h(t) \) is formed only on one side of the nanoparticle for half of the period of the oscillating field. Such an asymmetric motion would result in the generation of a dc electric field. Averaging over the period of oscillation, the voltage can be estimated as follows:

\[
U = \langle E(t)h(t)/2 \rangle = \frac{1}{2\pi} \int_0^{\pi} \Lambda E_0 \cos(\omega t) \frac{\varepsilon_0 \Lambda E_0 \cos(\omega t) \varepsilon_0 \Lambda E_0 \cos(\omega t)}{2\rho} dt = \frac{I \Lambda^2 l^3}{4eZe},
\]

where \( I \) is the light intensity.

Using \( Z = 1 \), and \( l \sim 0.5 \text{ nm} \), and the experimental value of \( P = 2 \text{ MW cm}^{-2} \), equation (1) yields \( U \sim 10^{-8} \Lambda^2 \text{ V} \).

Assuming the factor \( \Lambda \) to be on the order of \( \sim 10–30 \) at the resonance condition [26], the induced voltage would be anticipated to fall within the microvolt range, which is in line with the experimental observations.

The asymmetry of the boundary conditions can be associated with the direction of illumination and/or with the non-uniform nanoscale geometry. In figure 9(c), an SEM image
of the gold nanomesh fabricated under oblique angle deposition reveals a certain pattern: gold nanoparticles are seen inside the holes only on one side of the channels. In the experiment, such a structure shows unidirectional PLDE with the direction of the electron drag shown by the arrow in figure 9(c).

This simple approximation can describe both the polarity and magnitude of the induced emf. However, it cannot fully present the physical picture, taking into account that the estimated width of the positively charged layer, \( h \), is less or on the order of the lattice constant, \( l \). Alternative models can be based on the consideration of hot electron excitation and emission \([27]\) or thermal effects \([28]\) that may also exhibit such an asymmetry depending on the boundary conditions.

5. Conclusion

The plasmon-related PDE was studied in gold and silver films, gold and silver quasi-periodic nanomesh structures and arrays of Au-coated silicon nanopillars with well-defined periodicity. A strong enhancement of PLDE was observed in rough and nanostructured samples in comparison with nominally flat films. The maximum of PLDE was observed to occur at incident light excitation near the localized plasmon resonance condition. The angular dependence of the non-periodic systems demonstrated a sinusoidal dependence, while in the periodic arrays the dependence was more complicated. It was shown that the polarity and magnitude of the PLDE can be controlled with nanoscale geometry and can qualitatively be explained using a model based on the Drude approximation that accounts for non-symmetric boundary conditions.

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