New features in the surface plasmon induced photon drag effect in noble metal thin films

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Abstract
We study light–matter interactions leading to the generation of photon drag voltage under surface plasmon resonance conditions in noble metal thin films and observe important effects, which provide opportunity for condensed matter theorists to critically evaluate theoretical models. The drag voltage originates from a force that arises because of the transfer of momentum from incident light to electrons. This transfer of photon momentum leads to an electric current, which in turn results in the generation of drag voltage. The effect is particularly enhanced under surface plasmon resonance conditions and thereby reinforces the interaction between light and collective oscillations of the surface charges. We observe relatively high voltages with a nonlinear dependence on laser intensity. In disagreement with previous results, we do not observe a reversal in the sign of the voltage when the direction of the incident laser momentum is reversed. Qualitative analyses of the data show that the hydrodynamic model of laser-induced drag voltage does not work: the hydrodynamic model predicts voltages that are nine orders of magnitude lower than our measurements. However, there is reasonable consistency between the results of the measurements and numerical simulations.

1. Introduction

It has been known for decades that the transfer of photon momentum to carriers in semiconductors can generate a longitudinal electromotive force (emf) under open circuit conditions [1–8]. For example, Danishevskii et al studied in 1970 the dragging of free carriers by photons in direct interband transitions in semiconductors [9, 10]. These authors conducted their experiments on p-type germanium by using a high power (2 kW) Q-switched CO2 laser. They observed a reversal of the sign of the drag voltage with variation of sample temperature from ambient to liquid nitrogen temperature. In 1970 and 1971, Gibson et al also studied the transfer of momentum from a photon stream to free electrons and holes in p-type germanium [11, 12]. The major findings of this work included: (a) at room temperature the polarity corresponds to positive holes being driven by radiation pressure, (b) in some samples the polarity reverses upon cooling, and (c) the transition temperature of the photon drag effect in p-type germanium decreases markedly with increasing doping concentration. Grave et al published a theory of the photon drag effect in p-type germanium with a parabolic and anisotropic band structure approximation [13]. Wieck et al presented experimental evidence for a resonant photon drag effect in a 2D electron gas in GaAs quantum wells [14]. They argued that the drag voltage was induced by momentum transfer from 10 μm wavelength photons to the electrons resonantly matched in their intersubband energy to the photons by appropriate choice of the 8.2 nm well width. The voltage was reversed when the propagation direction of the photons was reversed. In the same context, Grinberg and Luryi have presented a theory of the photon drag effect in a 2D electron gas [15].

As is evident from the cited references, majority of the work on photon-induced drag voltage has been on semiconductors and essentially nothing on metals. Although plasmonic related effects have been investigated in metallic nanostructures. For example, Cai et al have used single-particle emission spectroscopy to resolve d-hole
relaxation in copper nanotubes [16, 17]. Similarly, Ostovar et al have studied the nanoparticle size dependence on photoluminescence and observed that small gold nanorods enhance intraband transitions [18]. The experimental work reported here utilizes the surface plasmon resonance (SPR) spectroscopy as one of the major techniques used. Therefore, we provide a brief description of this powerful optical spectroscopy technique. The SPR are excited either as surface plasmon polaritons (SPPs) propagating along metal/dielectric interface or as localized surface plasmons (LSPs) around metal nanoparticles (NPs) embedded in a dielectric [19]. The use of the surface plasmon resonance (SPR) for a variety of applications in physics, chemistry, biotechnology, electronics, environment, photonics, and the like has fueled tremendous interest in the development of SPR sensors. In this work, we used one of the widely used Kretschmann technique. In this technique, the base of a high-density prism is coated with silver or gold film. A polarized beam of laser strikes one of the faces of the prism at an angle higher than the critical angle for total internal reflection and reflected light intensity is measured as a function of the incident angle. As the surface plasmons are excited over the surface of the metal film, a loss in reflected intensity is observed. The ‘missing’ energy goes into the surface plasmons propagating along the sample surface. At resonance, the maximum loss of intensity occurs and the angle at which this happens is known as the resonance angle. From the practical point of view, it is often simpler to deposit the thin metal film over a substrate like Quartz and couple it to the prism with a refractive index matching fluid. We have utilized simple metal films as well as more sophisticated waveguide-coupled multilayer nanostructures [20–26]. Additional details are given later in this manuscript under experimental methods. It is only recently that photon drag effect has been observed in simple metals [27–37]. For example, Goff and Schaich presented a hydrodynamic theory of photon drag effect in simple metals (represented by jellium model) by adapting the hydrodynamic approach to second harmonic generation [27]. They consider that a laser beam of certain frequency illuminates the flat surface of a thick conductor in which only intraband transitions are important (semi-infinite jellium). The momentum transfer parallel to the surface of the sample amounts to a shear stress (light pressure). Under steady-state conditions the rate of transfer of momentum from the light to the electrons near the surface is balanced by resistive drag forces. The electrons gain momentum from absorbed light and lose momentum by resistive scattering, quickly settling into a steady drift parallel to the surface. Although a current is produced, no charge build-up occurs. The density of the parallel current varies with depth into the sample and vanishes beyond the penetration depth of the light field. Based on detailed calculations, Goff and Schaich show that the momentum balance arguments can provide exact results in special cases. However, they also observed that the predictions of this model can lead to qualitative wrong results in certain situations; a suitable variation of the electron relaxation rate as a function of depth into the metal can lead to a reversed direction for the surface-parallel current [27]. There have been several other notable works on the subject matter. For example, Ochiai studied the second harmonic generation and photon drag effect induced by an incident plane wave in doped graphene placed on a 2D diffraction grating, which acted as a plasmon coupler and dispersion modulator of the graphene plasmon. In this work, both the second harmonic generation and the photon drag effect were strongly enhanced by the excitation of the graphene plasmon polaritons [34].

As far as metals are concerned, a publication in 2000 by Goff and Schaich opens up the subject matter by their publication of the quantum mechanical self-consistent field theory of photon drag effect in simple metals [28]. This work shows a considerable sensitivity to the surface behavior of the responding electrons, with the strength of the surface contribution increased by two orders of magnitude compared to their hydrodynamic estimate. Noginova et al have observed surface plasmon polaritons-induced enhancement in the photon drag effect in thin silver films [31]. Similarly Kurosawa and Ishihara have studied the surface plasmon drag effect in a dielectrically modulated metallic thin film [38]. They investigated photo-induced voltage effect in Au film with a dielectric grating and observed strongly enhanced voltage when surface plasmon polaritons (SPPs) were excited. It was found that electrons in the Au film are driven to the propagation direction of SPPs due to the momentum transfer from SPPs to free electrons in the Au film. Recently Strait et al have conducted experiments on smooth gold film in vacuum and ambient conditions [36]. They discovered that the photon-drug photocurrent measured in vacuum was proportional to the momentum of light parallel to the surface of the sample, and the currents excited by using both polarizations of the incident light, as well as at both surfaces of the film were of the same sign. However, when the experiment was repeated in an ambient air environment, the p-wave photocurrent was dominated by molecular surface adsorbates. Additionally, the sign of the photon drag effect was found to be contrary to the prevailing intuitive model of direct momentum transfer to free electrons.

Since we have been studying the surface plasmon resonance in metals, it was a natural transition for us to investigate the effect in metal thin films [21, 23, 39]. In order to better understand the effect, we have completed a series of experiments on high-quality thin films of all three members of the noble metals as functions of the intensity, polarization and direction of the momentum of the incident photons, and the position of the incident laser beam between the two contacts on the sample used to measure the voltage. We supplement our experimental results by COMSOL [40]. simulations and evaluate detailed new results in terms of known theoretical models. The manuscript is organized as follows: (a) after an introduction in section-1, section-2
presents experimental details including the setup of initial conditions for COMSOL simulations, (b) section-3 presents results and discussion, and (c) section-4 concludes the work.

2. Experimental details

2.1. Thin film growth and characterization
High purity (≥ 99.999%) noble metals were used to deposit thin films on Quartz substrate by using thermal evaporation technique. As discussed below, the optimum thickness of each film (43 nm for Cu, 50 nm for Ag and 46 nm for Au) was determined by finite element method (FEM) based COMSOL simulations. The results of these simulations are in excellent agreement with those obtained by using MATLAB simulations and published elsewhere [41, 42]. An AJA International thermal evaporator was used in class-100 clean room NanoFab under 3 × 10⁻⁷ Torr pressure in the growth chamber, and at 35 °C–40 °C substrate temperature and 0.4 Å/sec deposition rate. Although films deposited at lower deposition rates are usually relatively uniform in thickness across the film area, contain cohesive grains and fewer defects, it does not really make much difference in results obtained in our experiments. We have noticed that for deposition rates in the 2 to 5 Å/sec range the results are not significantly different for the surface plasmon resonance or drag voltage measurements. Another point is that the level of control of the deposition rate with thermal evaporators, like ours in which the current is manually controlled, is subject to fluctuations resulting in deposition rate fluctuations. Therefore, it is not easy to lock down the current and deposition rate at a fixed deposition rate. This is a limitation of our deposition system.

Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM) were used to examine surface topography and microstructure of the films. The films were polycrystalline and densely packed with about a few hundred μm large crystallites. We did not explore the nature and the relative concentration of lattice defects [43]. Figure 1 shows a representative scan for a 50 nm thick Ag film.

2.2. Surface plasmon resonance spectroscopy

Since the realization of the plasmon excitations (PEs) in the 1950s, the subject matter continues to be of significant interest [44–47]. Pines and Bohm had suggested in the 1950s that energetic electrons, while passing through metallic foils, experience characteristic energy loss due to the excitation of ‘plasmons’ in the sea of the conduction electrons. The free electron approximation model of metals, in which the high density (≈10²³ cm⁻³) free electrons are treated as a liquid, supports plasma (‘volume’) oscillations propagating through the metal. The frequency of such plasmons is given by \( \omega_p = \sqrt{\frac{4\pi n e^2}{m_e}} \), where \( n \) is the density of the free electrons, \( e \) is the magnitude of the electronic charge, and \( m_e \) is the mass of the electron. The plasmon frequency is related to the frequency dependent dielectric function of the material through \( \varepsilon(\omega) = \left\{ 1 - \frac{\omega_p^2}{\omega^2} \right\} \). In the case of the free
electron metals, the energies of the volume plasmons range from approximately 8 eV for Li to about 16 eV for Al [48]. The subject matter of this review, however, is concerned with the surface plasmon excitations (SPEs), which are usually excited either as surface plasmon polaritons (SPPs) propagating along metal/dielectric interface or as localized surface plasmons (LSPs) around metal nanoparticles (NPs) embedded in a dielectric. The surface plasmon polaritons represent electromagnetic waves travelling along a metal/dielectric interface and having evanescent electromagnetic fields that decay exponentially normal to the interface. The localized surface plasmons are confined to metallic nanoparticles. The subject matter can be better understood by applying Maxwell’s equations to a set of two semi-infinite layers separated by a planar interface. A variety of mathematical software are available to obtain numerical solutions and simulations. We have utilized MATLAB and Finite Element Method of commercially available software COMSOL [19, 21, 39, 40].

2.3. Optical spectroscopy and four-probe resistivity measurements

Figure 2 shows the schematic of our optical spectroscopy system for simultaneous measurements of the surface plasmon resonance and induced electric voltage of thin film samples. The prism with a gold film deposited over its base is the ‘heart’ of the measurement system. A laser beam from a He-Ne laser passes through a beam splitter. The transmitted beam is polarized and steered by using mirror M1, which is mounted on a translational stage. A Keithley Nanovoltmeter is coupled to the four-point assembly and the measured voltage is sent to computer via LabView software.

Figure 2. Block diagram of the experimental set up for simultaneous measurements of the surface plasmon resonance and induced electric voltage of thin film samples. The prism with a gold film deposited over its base is the ‘heart’ of the measurement system. A laser beam from a He-Ne laser passes through a beam splitter. The transmitted beam is polarized and steered by using mirror M1, which is mounted on a translational stage. A Keithley Nanovoltmeter is coupled to the four-point assembly and the measured voltage is sent to computer via LabView software.
2.4. Numerical simulations

The surface plasmon resonance curves, and evanescent electric field of the surface plasmons were simulated by using the Finite Element Method in COMSOL multiphysics software [40, 56, 57]. Simulations were made by using unit cell of \( L \times W \times H \) where \( L = 632.8 \, \text{nm} \), \( W = 632.8 \, \text{nm} \) and \( H = 2.5 \times 632.8 \, \text{nm} \) of each sample. The size of the mesh elements varies from a minimum of 0.32500 nm (in metal) to a maximum of 126.60 nm (in air). Figure 4 shows representative results for the z-component (normal to the surface of the film) of the evanescent electric field and its distribution in 3-dimensions for a silver film. The SPR field is concentrated at the surface of the film and decays with increasing distance from the surface.

Results were also obtained for the SPR-induced photon drag effect, including the induced current density and evanescent electric field for the s- and p-polarizations of the laser beam. For the calculations of the current density, correction was made for a change in resistivity of the material under SPR conditions [24], and loss of the induced current density inside the film by using reasonable values of the skin depth (for example, for 50 nm thin Ag film skin depth is \( \sim 26 \, \text{nm} \)).
2.5. Determination of the optimum thickness of the film

The optimum thickness of each sample (43 nm for Cu, 50 nm for Ag and 46 nm for Au) is the thickness for which we observe the sharpest SPR curve with maximum loss in reflectivity at resonance. It was determined for each material by examining COMSOL simulations and measurements of the surface plasmon resonance curves [41, 42, 58]. A representative example is shown in figure 5 where the film thickness is varied from 44 nm to 56 nm. Film thickness of 50 nm provides the sharpest curve with maximum loss in reflectivity at resonance.

The best SPR curve (sharpest curve with almost 90% loss in reflectivity at resonance) is obtained for an optimum thickness of 50 nm for the Ag sample. In this figure, we show a set of SPR curves simulated and measured for different values of the film thickness. Obviously, the nature of the SPR curve changes with the thickness of the film. This has been amply demonstrated in our previous publications [19, 41, 42].

3. Results and discussion

3.1. Drag voltage data

Figure 6 shows the photon drag voltage and SPR data as a function of the incident angle for Cu, Ag, and Au films. The magnitude of the background voltage was obtained by turning off the incident laser and this was subtracted from the drag voltage data at each angle of incidence. The SPR data are in excellent agreement with previously published results [41]. It is striking that the drag voltage appears like a 'mirror-image' of the SPR data. The maximum drag voltage occurs at the surface plasmon resonance angle reinforcing the role of the surface plasmon polaritons in the transfer of the photon momentum to electrons on the surface of the material. It is also noteworthy that the magnitude of the drag voltage for all three noble metal films is much higher than that seen in previous experiments [59].

3.2. Effects of the laser intensity and direction of the photon momentum and polarization of incident light on drag voltage

We made a series of measurements of the drag voltage as functions of the intensity and direction of the momentum of the incident laser beam. Figure 7 shows effects of the laser intensity. Among the two sets of the data, one shows the actual measured values and the other shows values obtained from COMSOL simulations. Despite a small difference, there is good agreement between the trends of the intensity dependence of the drag voltage. The drag voltage changes with laser intensity quadratically indicating a second-order non-linear effect.

The effect of a change in the direction of the photon momentum on voltage is shown in figure 8. For positive values of the angle, the laser beam is incident upon the left face of the prism and the photon momentum is positive pointing from left to right along the surface of the film. In contrast to previously published data, we do not observe a sign reversal of the drag voltage when the laser beam is incident upon the sample from opposite faces of the prism [36]. Additionally, our data are not in agreement with the intuitive analogy of the radiation pressure on electrons in the sense that even when the direction of the laser momentum changes, the sign of the drag voltage does not change. The data are symmetrical about the vertical axis passing through zero angle. From a constant value of approximately 0.5 μV for small range of angles of about 10 degrees, drag voltage drops to about 0.3 μV. Thereafter the voltage rises sharply to about 1.4 μV at 35.3 degrees. Between 35.3 and 45 degrees, the drag voltage drops down to about 0.7 μV. It is also noteworthy the do not observe a drag
voltage when the polarization of the laser beam is switched from \( p \)-polarization to \( s \)-polarization. The drag voltage is, therefore, induced by the surface plasmon resonance. Figure 9 compares drag voltages for 43 nm Cu film of the \( s \) and \( p \) polarized light. Compared to a drag voltage of about 1.42 \( \mu \)V for \( p \)-polarized beam at resonance, it is only about 0.14 \( \mu \)V for \( s \)-polarized beam near resonance conditions.

3.3. Location of the laser spot and drag voltage: a local effect
The value of the drag voltage also depends on the location of the beam spot between the two contact points on the sample surface that are used to make voltage measurements. This is evident from the data shown in figure 10 for the (a) horizontal and (b) vertical locations of the laser spot in metal thin film. For the horizontal case, the zero beam-position corresponds to the mid-point in between the two contact points. Similarly, for the vertical case, the zero beam-position corresponds to the case in which the laser beam strikes upon, above and below negative terminal of pin contacts. Obviously, the effect is a local effect.

In the following sections, we discuss our data in terms of the known theoretical models.
3.4. The hydrodynamic model

Following Goff and Schaich, the hydrodynamic model considers that under steady-state conditions the rate of the transfer of the photon momentum must be balanced by resistive drag forces acting on the electrons near the surface of the sample \[27\]. The model provides the following equation for the time-averaged current per unit length flowing parallel to the surface,

\[
\langle dx J_l \rangle = \sigma_0 \left( \frac{S}{n_0 e c} \right) \sin \theta \cos \theta (1 - R),
\]

Where angular brackets denote a time average, \(J_l\) is the current per unit length flowing parallel to the surface, \(\sigma_0\) is the dc conductivity \((\sigma_0 = n_0 e^2 \gamma_b / m)\), \(n_0\) is the bulk electron density, \(e\) is the magnitude of the free electron charge, \(\gamma_b\) is the bulk relaxation time, \(m\) is the free electron mass, \(S\) is the incident Poynting flux along the beam direction, \(\theta\) is the angle at which the laser beam strikes the sample (angle of incidence), and \(R(\theta)\) is the reflection coefficient. In order to get an order-of-magnitude estimate, we use equation (1) and calculate angle dependent
time-averaged current per unit length flowing parallel to the surface of the Ag film. By using the measured value of the surface resistivity, the drag voltage can be calculated as a function of the incident angle $\theta$. Although equation (1) predicts more or less correct shape of the angle dependence of the drag voltage, the numerical values of the drag voltage, calculated by using reasonable values of other parameters in this equation, are orders of magnitude different from measured values. For example, if we use $S = \frac{1 mW}{\pi r^2}$, where 1 mW is the intensity of our laser beam and $r$ (~1 mm) is the radius of the beam spot (assumed circular, and without taking into account the loss in intensity of the laser beam as it propagates into the film), $\eta = 10^{-14}$ s, $n_b = 5.86x10^{28} m^{-3}$ for silver, $\mu = 1.6x10^{-19} C$, $m = 9.11x10^{-31} kg$, $c = 3x10^8 m/s$, $R(\theta)$ from measured reflectivity at $\theta = \theta_{SPR}$ and resistivity of silver film (0.09, 37 nΩ-m, respectively) the value of the drag voltage at the resonance angle turns out to be $-7.17 \times 10^{-15}$ V. It is nine orders of magnitude lower than the measured drag voltage of about $1.4 \times 10^{-8}$ V.

3.5. Optical rectification model

Another theory that has been used to calculate photon-induced drag voltage in nanostructures is the optical rectification theory. Kurosawa et al have presented a formalism for related optical rectification effect in metallic nanostructures with periodic boundary conditions [35, 60]. The optical rectification is a nonlinear process through which a quasi-DC polarization is generated in a nonlinear medium at the passage of an intense optical beam [61]. Kurosawa et al have applied the optical rectification theory to describe optical rectification voltage in a metallic thin film with a 700 nm period dielectric grating. The basis of the optical rectification process is the second order nonlinear polarization given by

$$P_i^{(2)} = \chi^{(2)}_{ik} E_i E_k + Q_{ijk} E_i \nabla_j E_k$$

(2)

Where $\chi^{(2)}_{ik}$ and $Q_{ijk}$ are second-order nonlinear susceptibility tensors of third and fourth rank, respectively. In this equation, the first term is the dipole term and it is associated with the space inversion symmetry of the material. The second term is a quadrupole term and it is associated with the symmetry of the electromagnetic fields. It can contribute significantly to second-harmonic generation, difference frequency generation in artificial structures, and optical rectification effect in metallic nanostructures [31, 35, 38, 66, 62]. These authors applied the optical rectification theory to calculate drag voltage for a metallic film with a dielectric grating of 700 nm period. Under the grating, a conductive metallic film of Au (40 nm)/Cr (3 nm) was grown over Quartz substrate. The drag voltages calculated by using this theory exhibited peaks and valleys corresponding to the surface plasmon resonance in agreement with experimental data.

3.6. Drag voltage from Finite Element Method simulations

As discussed above the FEM based simulation provides results for the strength of the evanescent field, field decay length, and net current density at the surface of the sample. In the case of the silver film, we obtain $9.64 \times 10^8$ A m$^{-2}$ for the volume averaged net current density, which amounts to a total current of $3.05 \times 10^{-5}$ A. By using the measured surface resistivity of the silver sample (37 nΩ-m) and simulation-provided total current of $3.05 \times 10^{-5}$ A, we obtain 2.56 μV for the drag voltage at the surface plasmon resonance angle. This is in reasonable agreement with the measured drag voltage of 1.4 μV.
4. Conclusions

We have presented new experimental data on the photon-induced drag voltage and its enhancement by the surface plasmon resonance in noble metal thin films. These data reinforce the intuitive picture of the transfer of momentum of the incident photons to the surface electrons as the basic mechanism by which a surface current appears and generates drag voltage. Detailed results are presented, which establish correlations between observed drag voltages and experimental parameters, like the direction of the photon momentum, intensity and polarization of the incident laser beam, and electronic properties of the material. It is for the first time that extremely high voltage has been observed for all three noble metals, and in disagreement with previous results, no sign reversal has been observed in voltage upon reversal of laser momentum. These data clearly establish the fact that the often-used hydrodynamic model does not work: its predictions are nine orders of magnitude lower than our measurements. However, the measurements are consistent with the predictions of the finite element method of COMSOL software. Hopefully, the work presented here will stimulate interest in the scientific community involved in the pursuit of related problems of light–matter interactions.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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