Magnetically dependent plasmon drag in profile modulated permalloy films

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Abstract. We explore coupling of plasmonic, electric and magnetic effects in profile modulated permalloy structures, which have both plasmonic properties and uniaxial magnetic anisotropy with an in-plane easy axis. Strong photocurrents, observed under plasmon resonance conditions, show a clear dependence on the magnetic field with a characteristic hysteresis. The effects are tentatively attributed to the inverse spin Hall effect.

Keywords: Plasmonics; Nanostructures; Magneto-optic systems; Inverse Spin Hall Effect

Introduction
The coupling of plasmonic and magnetic effects can bring many new possibilities and advantages, such as compact magneto-optical devices, fast magnetization switching, and nonreciprocal plasmonics [1]. The experiments in this direction are mostly focused on core-shell nanoparticles, which have a magnetic core (Co, Fe-Co, iron oxide) and a plasmonic shell (Au) or heterostructures of plasmonic and magnetic materials [2-7]. Significant enhancement of magneto-optical activity and an increase in photomagnetization under illumination with circularly polarized light are observed at plasmon resonance conditions, which clearly indicates the participation of surface plasmons in spin angular momentum (SAM) exchange between light and matter. The effects are explained with strongly enhanced electric fields at resonance conditions, and also discussed in terms of an enhanced inverse Faraday effect [8].

As shown in [9-11] surface plasmon polaritons (SPPs) propagating at the boundary between a dielectric and a metal have SAM associated with the rotation of the optical electric field polarization in the skin layer. Transfer of SAM to a material via spin orbital interaction can result in substantial spin polarization. In our work, we look for the signature of this process in photocurrent behavior and explore possible magneto-dependent electric response to plasmon excitation in systems, which combine plasmonic and magnetic properties. Strong enhancement of photocurrents in gold and silver structures observed at SPP resonance (plasmon drag effect) [13-18] is commonly discussed in terms of linear momentum exchange between light and matter [19-21], however, other factors (such as surface charges or highly nonlinear electron motion) can play a significant role [15, 21, 22]. If SAM transfer creates spin polarization, the overall picture of the photoinduced electric effects may be very different [23] from that in a purely non-magnetic case, bringing a new dimension to this research area.

Experimental
Our experimental structures are permalloy (Ni-Fe alloy with 80% Ni and 20% Fe) thin films with one-dimensional (1D) profile modulation, see Fig. 1 (a) for the schematics and Atomic Force Microscopy (AFM) image. The fabrication starts with obtaining polycarbonate grating substrates from disassembling commercial DVD-R discs by carefully taking out the polymer, plastic, silver and protective coating layers. Then, permalloy (Py) with 40 nm thickness is deposited on the prepared and precut DVD and glass substrates using e-beam evaporation. The thickness of the film is independently tested with a profilometer by measuring films simultaneously deposited on glass substrates. The AFM confirms that the Py/DVD systems have a periodicity, p, of 740 nm with a modulation height, h, of 60-80 nm. For optical characterization, the samples are illuminated with p-polarization with the grooves oriented perpendicular to the incidence plane. Reflectivity spectra show well-pronounced dips, which are ascribed to the SPP mode (1-st order SPP), following the equation [24],

\[ k_{\text{spp}} = 2\pi p + k_0 \sin \theta \]  (1)

where \( k_{\text{spp}} \) and \( k_0 \) are correspondingly the SPP and photon k vectors, and \( \theta \) is the angle of incidence.
The \( \omega(k) \) dispersion curve, Fig. 1 (c), derived from spectral positions of the dips at various angles is typical for a SPP [24]. The Q-factor of the resonance estimated from the dip width is relatively low, in the range of 3-10 depending on the wavelength; this is to be expected due to the high imaginary part of the optical permittivity in permalloy, \( \varepsilon = -7.76+15.9i \) [25]. Permalloy is a soft ferromagnetic with a very high magnetic susceptibility (up to 90000 [26, 27]). As shown in [28, 29], 1D-profile modulation of a magnetic film with submicron periodicity can produce uniaxial magnetic anisotropy with the in-plane anisotropy axis along the grooves. The magnetic properties of our structures are characterized with ferromagnetic resonance (FMR) method, which confirms in-plane uniaxial magnetic anisotropy with the anisotropy field \( H_a \) of \(~60-90\) Oe depending on a particular sample.

The experimental setup for photocurrent measurements is shown in Fig. 2 (a). The permalloy/DVD structure is prepared as a strip with a width of \( 3 \) mm and a length of \( 15 \) mm. The grooves are oriented perpendicular to the long side. Two electric contacts are attached to opposing ends of the strip. The sample is placed on a nonmagnetic stage and illuminated with the second harmonic of a Nd:YAG laser at 532 nm in p-polarization, with \(~10\) nm pulse duration and energy of \(~0.15\) mJ per pulse. The diameter of the illumination spot is slightly higher than the width of the sample. The voltage generated across the sample is measured with a Tektronix Digital Oscilloscope with 50 \( \Omega \) internal resistance. The magnetic field is supplied with an electromagnet. The direction of the magnetic field is parallel to the direction of the grooves.

We should note that voltages induced by the laser light illumination in the permalloy samples are significant, exceeding typical values in gold and silver surfaces obtained at the same or higher level of excitation power [12, 13, 18]. Just as with profile-modulated gold and silver systems [18], an enhancement of the photoinduced voltages is observed at incidence angles corresponding to the SPP excitation (plasmon drag effect). In this work, we restrict the discussion only to the magnetic dependence of the photoinduced voltages.

In the experiment shown in Figs. 2 (b, c), we set the illumination angle at the range of the SPP resonance \(~\pm 20^\circ\) in order to achieve the maximal electric signal, and compare the signals at the magnetic field \( H_0 = 85 \) Oe directed down (-z direction) and up (+z). In Fig 2 (b), the SPP is excited in the x direction, and the photoinduced electric signals have negative polarity, which corresponds to the drag of electrons in the x direction. As one can see, the peak
magnitudes of the photoinduced voltage are different for the opposite direction of the \( H \) field. The difference between the signals \( \Delta U = U(\pm H_0) - U(H_0) \) is shown in green. In Fig. 2 (c), the SPP is excited in the opposite direction, electrons drift in \(-x\) direction, but \( \Delta U \) has the same polarity and almost the same amplitude as in the previous case. At the SPP resonance conditions, \( \Delta U \) is higher (up to 2-3 times) in magnitude than it is off resonance, see Fig. 2 (d). The polarity of \( \Delta U \) always stays the same.

In Fig. 3, the photoinduced voltage is recorded with respect to an applied external magnetic field \( H \), which is slowly varied in the range between \( \pm 85 \text{ Oe} \). In Fig 3 (a), the \( H \) field first increases from negative to positive values (step 1-2). The voltage of \( \sim 3.7 \text{ mV} \) does not show any significant change in the magnitude until the applied magnetic field reaches \( 45 \text{ Oe} \). A steep drop in \( U \) from \( 3.7 \text{ mV} \) to \( 2.4 \text{ mV} \) is observed at \( 49 \text{ Oe} \). The voltage saturates at about \( 2.3 \text{ mV} \) with a further increase of the magnetic field. Reversing the magnetic field sweep (step 2-3) does not affect the voltage until the field reaches negative values of \( \sim -45 \text{ Oe} \). The voltage changes back to the original value \( 3.7 \text{ mV} \) at \( \sim -60 \text{ Oe} \) with a further increase in \( H \) in the \(-z\) direction, forming a full hysteresis loop.

In Fig. 3 (b), we test the effect of the field sweep when the SPP is excited in the opposite direction. Now we start from zero field and vary the field in the \(-z\) direction (step 1-2). Since the sample was previously exposed to negative fields, no switching is observed during this step. The voltage steeply changes from \( -5.4 \text{ mV} \) to \( -6.1 \text{ mV} \) only under positive fields during the field sweep in the \(+z\) direction (step 2-3) and stays at almost the same level during the step 3 - 4. A similar hysteresis behavior is observed in other samples as well, however the field where the switch in voltage occurs, slightly varies from sample to sample, see an example in Fig. 3 (c). Note that the magnitudes of the switching fields are comparable with the anisotropy fields \( H_a \) estimated from FMR characterization in our samples, indicating that the origin of the magnetic photocurrents is directly related to the magnetization of the sample.

![Fig 3. Hysteresis in photoinduced voltages under variation of the magnetic field. The field sweep direction is shown with the red arrows. Black arrows indicate the direction of the signal change. Dashed lines are a guide for the eye. Figs (a) and (b) correspond to different directions of illumination for the same sample. Data in (c) is obtained in a different sample. The data point shown in light green is obtained before introducing the field.](image)

In principle, the observed magnetodependence of the photoinduced voltage can be caused by a strong dependence of the electrical conductivity or optical permittivity on the magnetic field. After experimental tests of magnetoresistance and reflectivity measurements in a magnetic field, we exclude these factors from consideration as practically negligible.

**Discussion**

Summarizing our experimental observations, the magnetic part of the photoinduced voltage in profile-modulated permalloy films is related to spin polarization, and switches sign when the magnetization switches direction. It is enhanced by the SPP, however shows the same polarity at the opposite direction of SPP propagation. We believe that this effect is related to the Inverse Spin Hall Effect (ISHE) [30-36]. ISHE is due to spin-orbital interaction and allows one to obtain charge current from spin polarization current. Common demonstrations of this effect involve bilayer structures [30-34], where spin flow occurs via spin diffusion from a thin surface layer with non-equilibrium spin polarization created with microwave pumping at the ferromagnetic resonance frequency. The possibility to induce spin polarization via light illumination with circular polarization in doped semiconductors is discussed in [35,36]. Typical values of ISHE [30-36] are relatively low, in order of \( \mu \text{V} \).
Let us discuss our experiment, considering the scenario as following, see Fig. 4. Suppose the sample is magnetized down and assume a fast change of magnetization under laser pulse illumination (due to heating or SAM transfer via spin-orbital coupling) (Fig. 4 (a)). Since the light penetration depth in permalloy (estimated as ~13 nm from the optical data [25]) is much shorter than the thickness of the film, one can expect that the non-equilibrium magnetization strongly varies across the sample in the direction perpendicular to the film plane, Fig. 4 (b). A gradient of $\Delta M(y)$ causes the spin current [37]: electron spin up moves in the y direction, and spin down in the $-y$ direction. Due to ISHE, Fig. 4 (c), the spin current results in a charge current, $J$, in the x direction. In the case, when the sample is magnetized in the opposite direction, similar considerations yield the charge current in the ($-x$) direction. $U(-H_0) - U(H_0)$ is positive which corresponds to the experimental observations.

We should note that this scenario is only a tentative explanation for our findings, and more work is needed to elucidate the mechanism. The magneto-dependent voltages observed in our experiments are on the order of several mV, which are significantly higher than the typical values reported in Py [31]; this may be related to stronger gradients in spin polarization in our case, induced by high intensity illumination at plasmon resonance conditions.

An alternative scenario can be an anomalous spin Hall effect (AHE [31, 38], which would generate charge current in the $x$ direction when electrons are pushed by light in the $y$ direction. However, we believe that in our experiment this effect does not play a significant role, since it requires a unidirectional charge current across the film thickness during a relatively long period of time (at least, during the laser pulse), which is not expected in our geometry.

In conclusion, significant magnetic dependence of plasmon-enhanced photocurrents is observed in 1D profile modulated permalloy films, manifesting the coupling of plasmonic, electric and magnetic effects, which can open new opportunities in plasmonics and nanomagnetics.

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References
1. V. V. Temnov, G. Armelles, U. Woggon, D. Guzato, A. Cebollada, A. Garcia-Martin, J.-M. Garcia-Martin, T. Thomays, A. Leitenstorfer and R. Bratschitsch “Active magneto-plasmonics in hybrid metal–ferromagnet structures,” Nature Phot. 4, 107 (2010).
2. A. López-Ortega, M. Takahashi, S. Maenosono and P. Vavassori, “Plasmon induced magneto-optical enhancement in metallic Ag/FeCo core/shell nanoparticles synthesized by colloidal chemistry,” Nanoscale 10, 18672 (2018).
3. C. S. Levin, C. Hofmann, T.A. Ali, A. T. Kelly, E. Morosan, P. Nordlander, K. H. Whitmire, and N. J. Halas, “Magnetic-Plasmonic Core-Shell Nanoparticles,” ACS Nano. 3 1379 (2009).
4. K. Yang, C. Clavero, J. R. Skuza, M. Varela, and R. A. Lukaszew, “Surface plasmon resonance and magneto-optical enhancement on Au–Co nanocomposite thin films,” J. of Appl. Phys. 107, 103924 (2010)
5. I. Halagačka, M Vanwolleghem, K Postava, B Dagens, J. Pištora, “Coupled mode enhanced giant magnetoplasmonics transverse Kerr effect,” Opt Express 21, 2174 (2013).
6. P. Šrnoj, Y.-T. Chen, V. Vittur, M. D. Marquez and T. R. Lee, “Bimetallic Nanoparticles: Enhanced Magnetic and Optical Properties for Emerging Biological Applications,” Appl. Sci. 8, 1106 (2018).
7. D. O. Ignatyeva, G. A. Knyazev, P.O. Kapralov, G. Dietler, S. K. Sekatskii & V. I. Belotelov, “Magneto-optical plasmonic heterostructure with ultranarrow resonance for sensing applications,” Sci. Reports 6, 28077 (2016).
8. S. M. Hamidi, M. Razavinia, M. M. Tehranchi “Enhanced optically induced magnetization due to inverse Faraday effect in plasmonic nanostructures,” Opt. Commun. 338, 240-245 (2015).
9. K. Y. Bliokh, F. Nori, Transverse spin of a surface polaron. Phys. Rev. A 85, 061801(R) (2012).
10. T. Van Mechelen, Z. Jacob, “Universal spin-momentum locking of evanescent waves,” Optica 3, 118-126 (2016).
11. K.Y. Bliokh, A.Y. Bekshaev, F. Nori, “Optical momentum and angular momentum in complex media: from the Abraham–Minkowski debate to unusual properties of surface plasmon-polaritons,” New J. Phys. 19, 123014 (2017).
12. A. Vengurlekar, T. Ishihara, “Surface plasmon enhanced photon drag in metals,” Appl. Phys. Lett. 87, 091118 (2005).
13. N. Noginova, A. V. Yakim, J. Soimo, L. Gu, and M. A. Noginov, “Light-to-current and current-to-light coupling in plasmonic systems,” Phys. Rev. B 84, 035447 (2011).
14. H. Kurosawa, T. Ishihara, “Surface plasmon drag effect in a dielectrically modulated metallic thin film,” Opt. Express 20, 1561 (2012).
15. N. Noginova, V. Rono, F. J. Bezares and J. D. Caldwell, “Plasmon drag effect in metal nanostructures,” New J. Phys. 15, 113061 (2013).
16. H. Kurosawa, T. Ishihara, N. Ikeda, D. Tsuya, M. Ochiai, Y. Sugimoto, “Optical rectification effect due to surface plasmon polaritons at normal incidence in a nondiffraction regime,” Opt. Lett. 37, 2793 (2012).
17. M. Akbari, M. Onoda, M., and T. Ishihara, “Photo-induced voltage in nano-porous gold thin film,” Opt. Express 23, 823 (2015).
18. N. Noginova, M. LePain, V. Rono, S. Mashhadi, R. Hussain, and M. Durach. "Plasmonic pressure in profile-modulated and rough surfaces," New J. Phys. 18, 093036 (2016).
19. M. Durach, A. Rusina, M. I. Stockman, “GIant Surface-Plasmon-Induced Drag Effect in Metal Nanowires,” Phys. Rev. Lett. 103 (18), 186801 (2009).
20. M. Durach and N. Noginova, "On the nature of the plasmon drag effect," Phys. Rev. B 93, 161406 (2016).
21. H. Kurosawa, S. Ohno, and K. Nakayama, “Theory of the optical-rectification effect in metallic thin films with periodic modulation,” Phys. Rev. A 95, 033844 (2017).
22. J. H. Strait, G. Holland, W. Zhu, C. Zhang, B. R. Ilic, A. Agrawal, D. Pacifici, and H. J. Lezec, "Revisiting the Photon-Drag Effect in Metal Films," Phys. Rev. Lett. 123, 053903 (2019).
23. M. Durach, N Noginova, “Spin angular momentum transfer and plasmonic phenomena,” Phys. Rev. B 96 (19), 195411 (2017).
24. H. Raether, “Surface Plasmons on Smooth and Rough Surfaces and on Gratings,” Springer Tracts in Modern Physics 111. New York: Springer-Verlag 1988.
25. K. K. Tikušis, L. Beran, P. Cejpek, K. Uhliřová, J. Hamrle, M. Vaňatka, M. Urbánek, M. Veis, “Optical and magneto-optical properties of permalloy thin films in 0.7-6.4 eV photon energy range, “ Mater. Des. 114, 31-39 (2017).
26. H. D. Arnold and G. W. Elmen, "Permalloy, a new magnetic material of very high permeability," The Bell System Techn. J. 2, 101 (1923).
27. M. A. Akhter, D. J. Mapps, and Y. Q. Ma Tan, “Thickness and grain-size dependence of the coercivity in permalloy thin films,” J. of Appl. Phys. 81, 4122 (1997).
28. J. Berendt, J. M. Teixeira, A. García-García, M. Raposo, P. A. Ribeiro, J. Dubowik, G. N. Kakazeci, and D. S. Schmool, “Tunable magnetic anisotropy in permalloy thin films grown on holographic relief gratings,” Appl. Phys. Lett. 104, 082408 (2014).
29. S. A. Mollick, R. Singh, M. Kumar, S. Bhattacharyya and T.Som, “Strong uniaxial magnetic anisotropy in Co films on highly ordered grating-like nanopatterned Ge surfaces,” Nanotechn. 29 125302 (2018).
30. B. F. Miao, S. Y. Huang, D. Qu, and C. L. Chien, “Inverse Spin Hall Effect in a Ferromagnetic Metal,” Phys. Rev. Lett. 111, 066602 (2013).
31. H. Wang, C. Du, P. C. Hammel, F. Yang, “Spin current and inverse spin Hall effect in ferromagnetic metals probed by Y$_3$Fe$_5$O$_{12}$-based spin pumping,” Appl. Phys. Lett. 104, 202405 (2014).
32. P. Hyde, L. Bai, D. M. J. Kumar, B. W. Southern, C. M. Hu, S. Y. Huang, B. F. Miao, and C. L. Chien, “Electrical Detection of Direct and Alternating Spin Current Injected from a Ferromagnetic Insulator into a Ferromagnetic Metal,” Phys. Rev. B 89, 180404(R) (2014).
33. S. Y. Huang, X. Fan, D. Qu, Y. P. Chen, W. G. Wang, J. Wu, T. Y. Chen, J. Q. Xiao, and C. L. Chien, Transport Magnetic Proximity Effects in Platinum, Phys. Rev. Lett. 109, 107204 (2012).
34. K. Ando, J. Ieda, K. Sasage, S. Takahashi, S. Maekawa, and E. Saitoh, “Electric detection of spin wave resonance using inverse spin-Hall effect,” Appl. Phys. Lett. 94, 262505 (2009).
35. Jinling Yu, Xiaolin Zeng, Yumeng Wang, Lijia Xia, Shuying Cheng, Yonghai Chen, Yu Liu, Yunfeng Lai & Qiao Zheng, “Observation of Extrinsic Photo-Induced Inverse Spin Hall Effect in a GaAs/AlGaAs Two-Dimensional Electron Gas,” Nanoscale Res. Lett. 13, 320 (2018).

36. G. Isellaa, F. Bottegoni, A. Ferrari, M. Finazzi, and F. Ciccacci, “Photon energy dependence of photo-induced inverse spin-Hall effect in Pt/GaAs and Pt/Ge,” Appl. Phys. Lett. 106, 232402 (2015).

37. M. Vogel, R. Aßmann, P. Pirro, A. V. Chumak, B. Hillebrands & G. V. Freymann, “Control of Spin-Wave Propagation using Magnetization Gradients,” Sci. Rep. 8, 11099 (2018).

38. Y. Q. Zhang, N. Y. Sun, R. Shan, J. W. Zhang, S. M. Zhou, Z. Shi, and G. Y. Guo, “Anomalous Hall effect in epitaxial permalloy thin film,” J. of Appl. Phys. 114, 163714 (2013)