## Revisiting the Photon-Drag Effect in Thin Metal Films

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Current flow in metal films can be induced by the photon momentum carried by an obliquely-incident electromagnetic wave, a phenomenon known as photon drag<sup>1</sup>. The prevailing intuition for the sign of this current assume that the absorbed light transfers momentum to the free electrons of the metal, either directly or indirectly via plasmon excitation, ultimately generating electron flow in the direction of the in-plane incident photon momentum. However, the direction of this photon-drag (PD) current, has been reported to puzzlingly vary with polarization state, surface morphology and excitation of surface plasmons<sup>2 3</sup>. In particular, measurements to date have typically been carried out with the illuminated surface facing ambient air, yielding, for smooth films, PD currents of opposite sign for illumination with *s*- and *p*-polarization, respectively, where only the sign in the *p*-polarized case is consistent with the standard hypothesis of free-electrons pushed in the direction of the incident momentum (Figure 1).

Here, we demonstrate that for smooth metal films (including Au, Cu, and Ni-doped Ag) with illuminated surface facing vacuum, the PD current displays the same sign and nearly the same magnitude for both *s*- and *p*-polarized incident light, consistent with the expectation that light with different polarization states carries the same momentum (Figure 2). However, the shared sign of the observed current is always opposite to that implied by the intuitive model above, requiring reimagination of the microscopic processes of the light-metal interaction. We propose a new model for the PD phenomenon in which the bound electrons of the metal – far more numerous than the free electrons - are the primary recipients of the quantum momentum kicks from the incident light, leading to a net polarization of the metal to which the free electrons respond to yield a net PD current of the observed counterintuitive sign. The sign flip of the *p*-polarized signal upon subsequent exposure of the sample to air correlates with adsorption of ambient H<sub>2</sub>0 molecules, and is proposed to result from generation of an extra polarization field induced by the intrinsic dipole moment of the molecules upon Lorentz-force induced rotation.

Finally, we report on the design of coplanar-waveguide PD detectors based on ultrathin metal films deposited by ion-beam-assisted sputtering, for precision measurement of electromagnetic momentum in quantum optics applications.

<sup>&</sup>lt;sup>1</sup> Wegener M. Extreme Nonlinear Optics. Berlin: Springer, 2005.

<sup>&</sup>lt;sup>2</sup> Vengurlekar AS and Ishihara T. Surface plasmon enhanced photon drag in metal films. Appl Phys Lett 87: 091118, 2005.

<sup>&</sup>lt;sup>3</sup> Noginova N, Rono V, Bezares FJ, and Caldwell JD. Plasmon drag effect in metal nanostructures. New J Phys 15: 113061, 2013.



Figure 1: Photon-drag experiment (a) Experimental geometry. Using ~20 ps duration chirped pulses, centered at 80 nm wavelength, from a 1 kHz Ti:sapphire amplifier, an electrically contacted, ultra-smooth Au film, 25 nm thick, deposited on a glass substrate using ion-beam-assisted sputtering, is obliquely illuminated at angle  $\theta$ . Positive  $\theta$  is defined such that the in-plane photon momentum points toward the input of the oscilloscope. The device is mounted in a vacuum chamber, which is either vented with ambient air or baked and evacuated to  $<1x10^{-6}$  Pa. The optical pulses generate current pulses either parallel or anti-parallel to the in-plane incident photon momentum. The resulting voltage pulses, V(t) are measured across 50  $\Omega$  with a 300x, 1 GHz bandwidth amplifier and an 8 GHz bandwidth oscilloscope. (b) Representative measured waveforms for *s*- and *p*-polarized excitations, for  $\theta = +45^{\circ}$ , in ambient air. Though the prevailing intuition of photon drag would imply a negative voltage pulse for positive  $\theta$  (since it is expected that electrons would be "pushed" toward the amplifier input) only *p*-polarized light demonstrates this sign in air.



Figure 2: Effect of environment. Dependence of  $V_{\text{peak}}$  /A on  $\theta$ , where A is the fraction of absorbed power for each polarization at each angle, in (a) ambient air and (b) vacuum. Dotted lines: fit  $C \sin(\theta)$  (where C is a polarization-dependent, proportionality constant). Though the signals for s- and p-polarized light are of opposite sign when the sample is exposed to ambient air, the p-signal flips sign in vacuum, leading to similar behavior as a function of  $\theta$ , in both amplitude and sign, implying similar signal transduction for films in a pristine environment. However, the sign of this transduction is counterintuitive, since it implies net free-electron motion in the direction anti-parallel to the in-plane incident photon momentum.