THE IMPACT OF LASER POLARIZATION IN MULTIPHOTON PHOTOEMISSION FROM A COPPER CATHODE∗

R. K. Li, J. T. Moody, C. M. Scoby, H. L. To, M. Westfall, and P. Musumeci
Department of Physics and Astronomy, UCLA, Los Angeles, California, 90095, USA

Abstract

Multiphoton photoemission from a copper cathode has been recently demonstrated to be a simple and efficient method to generate high quality electron beams [1, 2]. To further improve this scheme to achieve higher charge yielding efficiency and lower intrinsic emittance, we explored the effects of laser polarization at oblique incidence. Charge yields of s and p polarization from coated and uncoated cathodes were measured. The vectorial photoelectric effect was observed on the uncoated cathode but much less evident on the coated one, suggesting that surface properties are critical to the vectorial effect and in general important in photoemission. The results not only are useful in the optimization of an rf photoinjector, but also allow deeper understanding of the photoemission physics.

INTRODUCTION

Excellent qualities of the electron beam generated by photocathode rf guns promise the successes of numerous recent applications, including x-ray free electron laser, inverse Compton scattering, and relativistic ultrafast electron diffraction. With remarkable advances in beam dynamics theory, simulation tools and experimentation over the past decades, beam qualities can be so well controlled during all acceleration, compression and transport stages, that it is believed the beam quality is now approaching a limit, the so-called ‘thermal’ or ‘intrinsic’ emittance, which is defined during the photoemission process. Recently more research interests [3, 4, 5] are focusing on photoemission for accelerator purposes, trying to better understand the physics, to choose or even engineer proper photocathodes and photon configurations, so as to deliver optimized emittance, quantum efficiency, temporal response, lifetime, vacuum requirement, etc., for various future applications.

Multiphoton photoemission from a copper cathode has been recently demonstrated to be a simple and efficient scheme for generating high brightness electron beams [1]. Traditionally, the output IR pulses from commonly used Ti:Sapphire ultrafast laser systems need to be frequency-tripled to UV and generate electrons through single-photon photoemission. Alternatively, one can directly send those IR pulses onto the cathode to generate electrons by n-photon photoemission, in which the charge yield scales as the nth power of laser intensity. When the laser pulses possess short duration, i.e. high intensity, n-photon photoemission can yield much more charge with the same amount of initial IR energies. Also, the photoelectrons emit promptly from the cathode and the initially ultrashort electron pulse dynamically evolves under its self-forces into a uniformly filled 3-D ellipsoid [2], whose emittance can be well compensated close to the intrinsic value.

A question that naturally follows is whether we can further improve the multiphoton photoemission scheme, in terms of charge yielding efficiency and intrinsic emittance. For single-photon photoemission, the vectorial photoelectric effect [6], that p polarized light generates more electrons than s for equal absorbed laser energy, has long been known. Though the theoretical explanation of this phenomenon remains debatable, from practical point-of-view sending p polarized laser at an optimal angle may yield the highest amount of charge for a given laser energy. Moreover, laser polarization may modify the intrinsic emittance of the photoelectron beam. Quantum mechanical (QM) calculations [7, 8, 9] indicate that photoelectrons only emit from the surface of the material, and roughly follow a $\cos^2 \theta$ distribution around the laser electric field component normal to the cathode. In contrast, phenomenological treatments like the three-step model [10], which does not include the effect of laser polarization, suggests that photoelectrons originate from the bulk of the material and distribute isotropically in the half-space outside the cathode. If photoelectrons actually can be grouped as surface and bulk emitted ones and follow different angular distributions, it is predicted [11] that one can minimize the beam emittance using an optimal laser polarization and incidence angle.

In this paper, we report on the measured charge yield for s and p polarized IR laser pulses at oblique incidence by multiphoton photoemission from coated and uncoated copper cathodes. For both cathodes p polarization is more efficient than s, but for the coated one the enhancement of p over s is only due to reflectivity. We discuss possible physical explanations of above observations.

EXPERIMENT RESULTS

The experiment took place at the UCLA Pegasus laboratory. IR laser pulses illuminated the copper cathode of an S-band 1.6 cell photocathode rf gun with a 72.5° incidence angle through the laser port on the half-cell. The laser spot-size on the cathode was defined by an iris before the laser port, and a cylindrical lens was inserted to compensate for the ellipticity due to oblique incidence.

The coated cathode was prepared by off-axis single-point diamond turning and a MgF2 antireflective coating...
for UV. The uncoated cathode was polished with 9 μm to 0.25 μm monocrystalline diamond paste and cleaned in hexane ultrasonic bath. The reflectivities of \( s \) and \( p \) polarized lasers at 72.5° angle of incidence on both cathodes are listed in Table 1.

Table 1: Measured reflectivity \( R \) of the coated and uncoated cathode for \( s \) and \( p \) laser polarization.

<table>
<thead>
<tr>
<th>cathode</th>
<th>( R_s )</th>
<th>( R_p )</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgF(_2) coated</td>
<td>63.8%</td>
<td>48.0%</td>
</tr>
<tr>
<td>uncoated</td>
<td>87.2%</td>
<td>71.1%</td>
</tr>
</tbody>
</table>

Fig. 1 (a) and (b) show the charge yield density versus the incident laser intensity \( I \) for the coated and uncoated cathode, respectively. Each data point reflects the measurements of a single laser pulse. The charge of the electron bunch was measured by a Faraday cup at the end of the beamline. The laser pulse energy was monitored by a calibrated photodiode on a split laser path out of the main beam. The gradient and injection phase of the photocathode rf gun were kept constant at \( E_0 = 55 \text{ MV/m} \) and \( \phi = 30° \). The laser pulse length \( \tau \) was minimized by adjusting the compressor and measured off-line with a polarization gating based autocorrelator, so small differences in day-by-day laser operation were possible. An estimated laser pulse length of 100 fs FWHM is used for all the curves. In Fig. 1 (c) and (d) we plot the charge yield density as a function of absorbed laser intensity \( I(1-R) \) using the measured reflectivity data.

**DISCUSSION**

**Observation I:** As shown in Fig. 1, for both cathodes and both laser polarizations, the charge yield density increases as the 3rd power of the incident or absorbed laser intensity, provided the image-charge induced electric field is much smaller than the rf field \( E_0 \sin \phi \).

This observation can be explained by either phenomenological models like the generalized Fowler-Dubridge (FD) theory [12], or QM calculations such as Ref. [7, 8, 9].

The generalized FD theory states that the photocurrent density of a \( n \)-photon photoemission process is

\[
J_n^{\text{FD}} \propto a_n I^n (1-R)^n,
\]

where \( a_n \) describes the likelihood of the \( n \)-photon process. \( a_n \) is difficult to calculate accurately, thus it is basically an empirical parameter. The reflectivity \( R \) essentially remains constant over the intensity range of interest for photoemission, which is below the damage threshold at \(~100 \text{ GW/cm}^2\) for sub-ps laser pulses.

QM calculations treat photoemission as a one-step process by solving the Schrödinger equation with a Hamiltonian containing the incident laser field and the potential barrier of the cathode surface. It predicts that the photoelectrons are emitted, for metal at least, only from the surface. The photocurrent density scales as

\[
J_n^{\text{QM}} \propto E_{\perp}^{2n},
\]

where \( E_{\perp} \) is the component of the electric field normal to the surface at the metal side of metal-vacuum or metal-dielectric interface. \( E_{\perp} \) is difficult to measure experimentally and strongly depends on the material properties, surface profile and laser polarization. For a given cathode and laser configuration the integration of \( E_{\perp}^2 \) over the illuminated area is proportional to the absorbed laser intensity, thus it is natural to expect Observation I.

**Observation II:** On both cathodes \( p \) polarized lasers yield more electrons than \( s \) with equal incident intensity. After taking into account the difference in reflectivity, with equal absorbed laser intensity, the charge yield by \( p \) and \( s \) from the coated cathode are almost the same or comparable to the experiment uncertainty; however from the uncoated cathode \( p \) still generates 70% more photoelectrons.

The vectorial photoelectric effect has been observed with several metal and semiconductor materials, including Cu [13, 14, 15], Mg [16, 17], Mo [18], Au [19], W [20] and...
Si [6], etc. Regarding the measurements on copper cathode photoinjectors [21], several groups reported the charge yield variation due to laser polarization, although in some cases only the incident laser energy was concerned rather than the absorbed one.

Theoretical explanation of the extra charge yield per p over s for equal absorbed intensity lies beyond the FD theory or the three-step model. The theories of surface photoemission predict that the change of the EM vector potential A across the metal-dielectric boundary, \( |\nabla \cdot A| = |A_\perp(\epsilon_1 - \epsilon_2)|/d \), will make extra contributions to photoemission, where \( \epsilon_{1,2} \) are the dielectric constants of the materials and d is the distance across the boundary. Similarly to Ref. [20], we attempt to evaluate the relative significance of surface photoemission due to \( |\nabla \cdot A| \). We assume that s polarization generates photoelectrons only through bulk emission while p polarization causes both bulk and surface photoemission. The macroscopic normal electric field component \( E_{p\perp} \) is used as an indication of \( |\nabla \cdot A| \) since \( |A_\perp| \propto |E_\perp| \). A parameter \( r \) represents the extra contribution of the surface effect. The charge yield ratio for equal absorbed laser intensity (\( E_{p0}^2 = E_{s0}^2 \)) is

\[
\frac{Y_p(E_{p0})}{Y_s(E_{s0})} = \frac{E_{p0}^6 + r E_{p\perp}^6}{E_{s0}^6} = 1 + r \sin^6 \theta, \tag{3}
\]

where \( \theta \) is the angle of incidence. For the uncoated and coated cathodes, \( r \) is 0.9 and -0.2, respectively.

At present we do not have a quantitative model including the effects of surface roughness, coating and possible contamination, but it is fair to conclude that surface effect is observed on the uncoated cathode, however is much less evident on the coated one. The possible reasons for the difference include the depressed \( |\nabla \cdot A| \) across the metal-dielectric boundary compared to the metal-vacuum one and roughness induced surface plasmons which can efficiently use the absorbed energy from s and p polarization.

**Observation III:** The reflectivity of the uncoated cathode is evidently different from that reported in Ref. [22], which is measured on an evaporated thin film. We measured several other cathodes and observed that the reflectivity decreases with increased surface roughness, although the effects of surface oxidation and contamination can not be ruled out.

Surface properties not only affect the reflectivity but also determine the EM energy distribution across the boundary. QM calculations predict that photoelectrons originate from a region only a few atomic layers below the surface; in the three-step model the e-e scattering length of excited electrons is also only \( \sim 2 \) nm: both numbers are much smaller than the optical penetration depth which is \( \sim 10 \) nm or longer. We argue that the photoelectron yield is only influenced by the EM energy density within a few nm from the surface, rather than the total absorbed optical energy which may extend over tens of nm. In this light the charge yield can be enhanced by concentrating the EM energy density at the metal side of metal-vacuum or metal-dielectric boundary, e.g. by exciting surface plasmons [23, 24, 25].

**CONCLUSION**

In summary, we measured the charge yield of multiphoton photoemission using 800 nm s and p polarized ultra-short laser pulses from uncoated and coated copper cathodes. We observed the vectorial photoelectric effect on the uncoated cathode in which p polarization generates more photoelectrons than s with equal absorbed laser intensity, but the effect is much depressed on the coated cathode. The results indicate that surface properties are critical for the vectorial photoelectric effect, and in general, important in photoemission. Systematic correlation measurements of the total yield, angular distribution and energy spectrum of photoelectrons from a clean surface with controlled roughness or coated surface are necessary to fully understand the photoemission physics from the simple and widely used copper, so as to guide us to optimize the photocathodes for future accelerator applications.

**REFERENCES**

[25] H. Padmore, at the PPP Workshop, see Ref. [4].