Evidence of vectorial photoelectric effect on Copper

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Abstract

Quantum Efficiency (QE) measurements of single photon photoemission from a Cu(111) single crystal and a Cu polycrystal photocathodes, irradiated by 150 fs-6.28 eV laser pulses, are reported over a broad range of incidence angle, both in s and p polarizations. The maximum QE ($\simeq 4\times10^{-4}$) for polycrystalline Cu is obtained in p polarization at an angle of incidence $\theta = 65°$. We observe a QE enhancement in p polarization which can not be explained in terms of optical absorption, a phenomenon known as vectorial photoelectric effect. Issues concerning surface roughness and symmetry considerations are addressed. An explanation in terms of non local conductivity tensor is proposed.

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The advent of the 4th generation free electron lasers (FEL) sources [1–3] triggered several important technical questions. A fundamental issue regards the photocathode material for the laser-driven photoinjector devices, to obtain short electron bunches with high charge density and low emittance. Metal photocathodes are good candidates, having a high reliability, long lifetime and a fast time response (1-10 fs). However, two major drawbacks limit their usefulness, the small quantum efficiency (QE) and the high work function (Φ), requiring light source in the ultraviolet (UV) for efficient linear photoemission.

In this Letter we study the experimental conditions to maximize the QE of Cu photocathodes using UV short laser pulses from the quadrupled output of an amplified Ti:Sapphire laser. The QE for linear photoemission in the femtosecond regime is measured as a function of the angle of incidence θ in the angular range $-55° \leq \theta \leq +80°$, both in s and p polarizations. The maximum quantum efficiency $Y \simeq 4 \times 10^{-4}$, obtained with p polarization at $\theta = 65°$, is four times the value at normal incidence.

The QE dependence on angle of incidence and light polarization is a long standing problem [4–8] that largely remains to be understood. Our data are well fitted by a phenomenological model [6] that keeps into account only light absorption, without any explanation in terms of microscopic quantum physics. A justification of the phenomenological model based on the calculations of the conductivity tensor for a jellium model is proposed.

The photoemission from a polycrystalline Cu sample and a Cu(111) single crystal is investigated with 150 fs laser pulses with a photon energy of 6.28 eV, obtained by two successive doubling processes of the Ti:Sapphire fundamental frequency ($h\nu = 1.57$ eV) in β-barium-borate (BBO) crystals. The second doubling process is obtained out phase-matching in a thin (200 μm) BBO crystal. The fourth harmonic is selected by dispersing the doubling crystals output with a MgF_2 prism, with minimal temporal and pulse front tilt distortions.

We do not use a more efficient third harmonic conversion to obtain linear photoemission from Cu ($3h\nu = 4.71$ eV, $\Phi = 4.65$ eV for polycrystalline Cu [9]) because of the onset of multiphoton regime upon a work function increase due to sample contamination. Moreover, an effective laser-induced oxide removal and contaminants chemical-bond breaking obtained with UV short laser pulses [10, 11] improves with shorter wavelengths [10]. Working with a 6.28 eV photon energy should thus help to increase the duty time of machines based on Cu photocathodes.

The quantum efficiency Y is the ratio between the number of photoemitted electrons, obtained from the photocurrent measured from the sample with a Keithley 6485 Picoammeter, and the number of incident photons, detected measuring on a Tektronix TDS3054B digital oscilloscope the output of a Hamamatsu R928 photomultiplier tube. The measurements are performed with the Cu photocathodes kept in a ultrahigh vacuum chamber with a base pressure of 2×10^{-10} mbar at room temperature. During the total yield measurements, the photoemission spectrum is acquired using a time of flight spectrometer in order to measure the sample work function and monitor possible onset of sample contaminations and space charge effects. The samples are cleaned by cycles of Ar^+ sputtering followed by annealing at 500°C. This procedure is continued until the proper value of the measured work function (4.65 eV for the polycrystal and 4.94 eV for the single crystal) is obtained. In these conditions a clear low energy electron diffraction (LEED) pattern for the Cu(111) sample is obtained. The laser peak intensity on the target is $I \simeq 5 \times 10^5$ W/cm^2.

The QE measured for both samples are reported in Fig. 1. An enhancement of the QE is evident for p polarization as compared to what would be expected taking into account only the electromagnetic absorption process. The maximum QE do not occur at the pseudo-Brewster angle of incidence $\theta_B = 57°$ [12](see Fig. 1), where there is maximum absorption, but is shifted by $\sim 8°$ toward the normal.

Our experimental results can be rationalized in the frame of a phenomenological model proposed in Ref. 6. The electric field transmitted inside the sample can be written as $\mathbf{E} = \mathbf{E}_p + \mathbf{E}_s = \mathbf{E}_\parallel + \mathbf{E}_\perp$, where $\mathbf{E}_p$ and $\mathbf{E}_s$ are the p and s polarized field components respectively, $\mathbf{E}_\parallel = \mathbf{E}_{p\parallel} + \mathbf{E}_s$ and $\mathbf{E}_\perp = \mathbf{E}_{p\perp}$ are the components parallel and perpendicular to the surface respectively. The electric field vector components are defined in Fig. 2. The QE, normalized with respect to its value at normal incidence $Y(0)$, is:

$$\frac{Y(\theta)}{Y(0)} = \frac{\varepsilon_\parallel(\theta)}{\varepsilon_\parallel(0)} + r\frac{\varepsilon_\perp(\theta)}{\varepsilon_\parallel(0)}, \tag{1}$$

where $\varepsilon_\perp = \varepsilon_{p\perp}$ and $\varepsilon_\parallel = \varepsilon_{p\parallel} + \varepsilon_s$ are the electromagnetic energies inside the sample due to the fields components indicated by the suffixes. A value $r = 1$ means that photoemission is proportional to the absorbed intensity, whereas $r > 1$ implies that $\mathbf{E}_\perp$ is more efficient than $\mathbf{E}_\parallel$ in producing photoelectrons. Eq. 1 specialized for p polarization is:

$$\frac{Y_p(\theta)}{Y_p(0)} = \frac{\varepsilon_{p\parallel}(\theta)}{\varepsilon_{p\parallel}(0)} + r\frac{\varepsilon_{p\perp}(\theta)}{\varepsilon_{p\parallel}(0)}, \tag{2}$$

whereas for s polarization ($\mathbf{E}_\perp = 0$):

$$\frac{Y_s(\theta)}{Y_s(0)} = \frac{\varepsilon_s(\theta)}{\varepsilon_s(0)}. \tag{3}$$

Once the electromagnetic energies $\varepsilon_{p\parallel}(\theta)$, $\varepsilon_{p\perp}(\theta)$ and $\varepsilon_s(\theta)$ are calculated from classical electrodynamics, assuming volume absorption as in Refs. 6, 13, the parameter r is obtained fitting the experimental data for p polarization with Eq. 2. The best fit values are $r = 13$ for the polycrystalline Cu and $r = 9$ for the Cu(111) single crystal (see Fig. 1). The QE dependence expected on the basis of Fresnel laws only, setting $r = 1$, is also reported as a dashed line in Fig. 1. The data for s polarization are in agreement with Eq. 3.

At the light of our data, it is important to investigate the physical mechanisms that enhances the photoelectron yield due to $\mathbf{E}_\perp$ over $\mathbf{E}_\parallel$.

The crystalline symmetry, important when dealing with polarization dependent photoemission, play no role in the present experiment. The photoemission process due to $\mathbf{E}_\perp$ is about 10 times more effective than $\mathbf{E}_\parallel$ both in the Cu(111) single crystal, where symmetry considerations could apply, and in the polycrystalline Cu, where any symmetry-related contribution is cancelled by the random orientations of the single crystals domains composing the sample.

Photoemission enhancement due to surface roughness has been recently investigated [14–16]. In the present case surface roughness enhancement can be ruled out. Several atomic force microscopy (AFM) scans of the samples surface, with sizes ranging from 1×1 μm^2 to 60×60 μm^2, give values of the root mean squared roughness $h_{rms} \sim 20$ nm for the Cu polycrystal and $h_{rms} \sim 2$ nm for the Cu(111) single crystal, see Fig. 3. The observed vectorial photoelectric effect is comparable on both samples, despite their surface roughnesses differ by an order of magnitude. The comparative study of the single crystal Cu and polycrystalline Cu cathodes allows to clarify that our experiment is not dependent on sample morphology.

Therefore, we seek for an explanation in terms of a more general mechanism. Solutions of the Maxwell equations on an ideal jellium-vacuum interface for an impinging plane electromagnetic wave of frequency ω, evidence an electromagnetic field spatially varying on the length scale of ~ 1 Å on the jellium side [17]. The spatially varying electromagnetic field is due to the non local character of the conductivity tensor. This is calculated using free-electron like wave functions, so it does not depend on the symmetry of the crystal. The matrix element entering the differential cross-section for photoemission is composed of two

terms. The first is the usual electric dipole contribution, the second is due to the rapidly varying electric field. The second term prevails for $\omega < \omega_p$, where ω_p is the plasma frequency, and leads to an enhancement of the photocurrent for the electric field components perpendicular to the sample surface [18, 19]. In the present experiment, $\hbar\omega = 6.28$ eV and $\hbar\omega_p \sim 19$ eV [20]. This mechanism explains an enhancement of the QE for p polarized incident radiation while not affecting the results for s polarized light. Furthermore, it does not depend on surface roughness or a particular symmetry of the crystal. We therefore propose it as the main microscopic mechanism to explain our experimental evidences.

In this Letter quantum efficiency measurements on Cu photocathodes, irradiated with 150 fs laser pulses at 6.28 eV, are reported over a broad range of incident angles in both s and p polarizations. A QE enhancement is found for light with electric field perpendicular to the sample's surface, showing a vectorial photoelectric effect. The maximum value of quantum efficiency $Y \simeq 4 \times 10^{-4}$ is four times bigger than the QE at normal incidence and is achieved with p polarized light impinging on the sample at an incidence angle of $\theta = 65°$. Investigation of both a Cu(111) single crystal and a Cu polycrystal allows us to rule out a microscopic processes based on symmetry considerations and surface roughness to explain our data. An explanation in terms of a rapidly varying effective field, due to the non local character of the conductivity tensor, is suggested.

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Figures

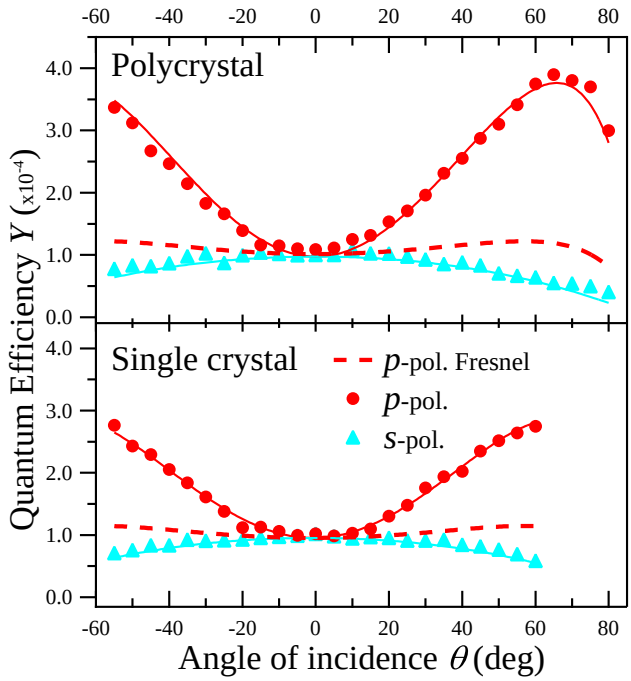

FIG. 1: Measurements of quantum efficiency dependence on the angle of incidence θ for a Cu polycrystal and a Cu(111) single crystal for p (circles) and s (triangles) polarized light. Fits, based on Eq. 1, are reported as solid lines. The dashed lines are calculated taking into account Fresnel absorption only.

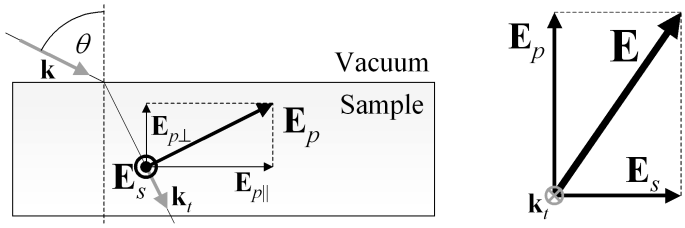

FIG. 2: Representation of incidence angle θ, wave vectors k and k_t for incident and transmitted light and field components addressed in the text. A real index of refraction n is assumed for the present figure.

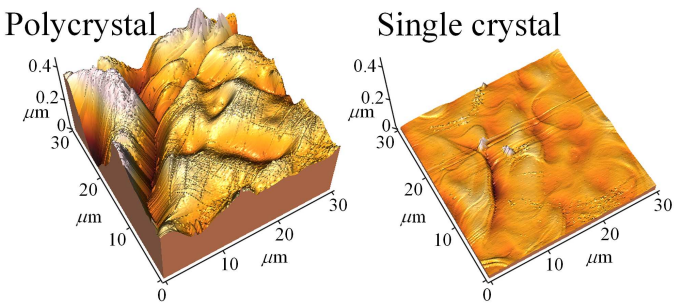

FIG. 3: Atomic Force Microscopy images of the two samples' surfaces. Measured route mean squared roughness is 20 nm for the Cu polycrystal and 2 nm for the Cu(111) single crystal.