PHOTOCATHODE R&D PROGRAM AT LBNL*

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Abstract

The work of the photocathode R&D program at LBNL is presented. The quantum efficiency (QE) of Cu(111) is measured for different impinging light angles with photon energies just above the work function. We observe that the vectorial photoelectric effect, an enhancement of the quantum efficiency due to illumination of light with an electric vector perpendicular to the sample surface, is stronger in the more surface sensitive regime. This can be explained by a contribution to photoemission due to the variation of the electromagnetic potential at the surface. The contributions of bulk and surface electrons can then be determined. Angle-resolved photoemission data let us obtain the dispersion relation of the surface state, which in turn allows us to determine the thermal emittance of the electrons emitted from the surface.

INTRODUCTION

The advances in diverse fields such as electron microscopy and the free electron laser drive demand for high brightness electron sources [1]. The photocathode R&D program at LBNL was established to explore ways of producing new generation of photocathode that can meet the demand. Our program is designed to study in detail the yield and the energy-momentum dispersion relation of the emitted electrons using angle-resolved photoemission spectroscopy (ARPES), as well as to develop new photocathode using nanotechnology. The capability of fully characterizing the photocathode in the ideal setting allows us to find out the physical limit of the performance of the material, as well as pointing to the direction of making better photocathode. In addition, the gap between the ideal setting and the realistic situation in a gun can be simulated to a certain extent in a controlled manner.

EXPERIMENTS ON CU(111)

The first set of experiments were carried out in Brescia, Italy on Cu(111). New work is carried out using a similar system in the photocathodes lab at the ALS. Although copper has been widely used in RF photoguns [2], there are still a lot of questions remain unanswered, such as the origin of the emitted electrons and the lower bound of the transverse thermal emittance. Furthermore, Cu(111) was chosen as a sample due to its robust nature and its well known and experimentally verified band structure [3].

An amplified Ti:Sapphire laser was used as the light source in this experiment, providing 150~fs 790~nm pulses with an average light power of 500~mW at 1~kHz repetition rate. The output was split into two beams: the first pumping a parametric amplifier that provided tunability in the near infrared, the second undergoing a process of third harmonic generation obtained with two stages of sum frequency generation in type I BBO crystals. After a delay line that provides temporal coincidence, the beams converge on a third crystal for sum frequency generation providing the desired wavelengths. The sample total current on a picoammeter and the light intensity of a beam reflection on a calibrated photodiode were measured to provide the experimental data.

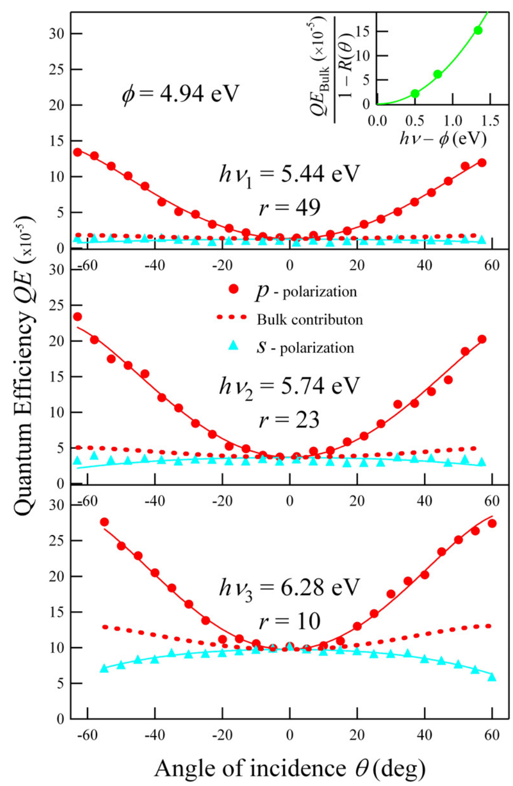

Figure 1: Quantum efficiency data as a function of the incident light angle θ for the three different photon energies are fitted using Eq. (1), (2); the bottom panel shows data from Ref. [4]. The red dotted lines represent the prediction for the bulk contribution, proportional to the absorbed part (1-R(θ)) of light energy. The top right panel shows the bulk quantum efficiency fitted by the Fowler law Eq. (3).

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The experiment was performed in an ultrahigh vacuum chamber with the base pressure of 2e-10 mbar at room temperature on a Cu(111) crystal polished with cycles of Ar^+ sputtering at 1 keV kinetic energy and annealing at 750 K. The sample cleanliness was tested online by acquiring photoemission spectra through a time of flight analyzer and checking the sharpness of the L gap surface state and the work function value.

The quantum efficiency was measured as a function of the incidence angle θ of the impinging photons in the range $-63^\circ < \theta < 57^\circ$ with 5 steps ($\theta = 0^\circ$ indicates normal incidence) for two different values of the photon energy $h\nu_1$ = 5.44 eV and $h\nu_2$ = 5.74 eV; data are compared to results of Ref. [4], obtained with $h\nu_3$ = 6.28 eV.

The most widely applied theoretical model is the 3-step model [5], which assumes that the yield depends on the total absorbed photon energy. This is proportional to the incident light intensity and to (1-$R(\theta)$), where $R(\theta)$ is the reflectivity calculated from the Fresnel laws [5]. Yet this prediction is not confirmed by Cu(111) data obtained for *p* polarized light. From the phenomenological point of view, the dependence of the quantum efficiency on the *s* and *p* polarized light at a given incidence angle can be expressed as

$$\frac{QE_s(\theta)}{QE(0)} = \frac{\varepsilon_s(\theta)}{\varepsilon_s(0)} \tag{1}$$

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

where $QE(0)$ and r are fit parameters. The parameter r is a measure of the ratio between the effectiveness of perpendicular and parallel light fields [6][4]. It is clear from Fig. 1 that r increases as the photon energy decreases, which indicates that the contribution of the surface becomes more prominent (for details, see ref. [7]).

Another feature of Fig. 1 is that the bulk emission obeys the Fowler law, which is

$$\frac{QE_{bulk}(\theta)}{1-R(\theta)} = \frac{QE_{bulk}(0)}{1-R(0)} \propto (h\upsilon - \phi)^2 . \tag{3}$$

Yet the surface emission violates the Fowler law.

In addition to yield measurement, angle resolved time-of-flight spectrometer was used to measure the energy and momentum distribution of the photoelectrons. The data is presented in Fig. 2, which shows that, even at the photon energy of 5.71 eV, virtually all photoelectrons are from a surface state [8]. From the set of data, we obtain the effective mass of the electron, which is $0.45m_e$, where m_e is the free electron rest mass. This is in excellent agreement with previous results, which is 0.46 m_e [8].

With the knowledge that most photoelectrons are from the surface state and that the energy-momentum dispersion is known, we can compute the transverse emittance of the emitted electrons. Here we adopt the normalized emittance, which is definition as

$$\varepsilon_{x,rms} = x_{rms} p_{rms} /(m_e c) , \tag{4}$$

where c is the speed of light. For uniform emission, we have

$$x_{rms} = \frac{1}{2}R , \tag{5}$$

where R is radius of the emitting area. For free electron like dispersion relation, it is reasonable to assume that $d^2N/dp_x dp_y$ is constant. Hence we get

$$p_{x,rms} = \frac{1}{2}P_r . \tag{6}$$

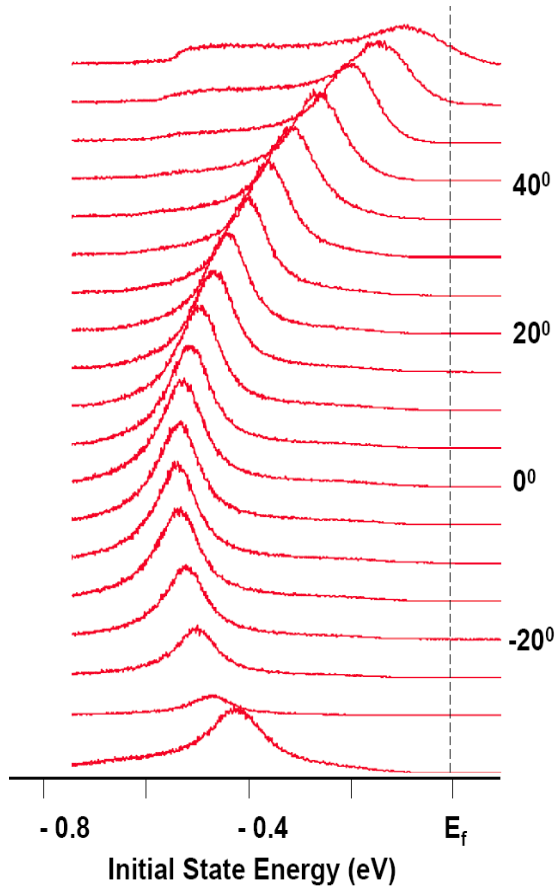

Figure 2: Energy spectrum of the photoelectrons for various angles of the electrons with respect to the norm of the surface. The photon energy is 5.71 eV. The ratio of the effective mass and the rest mass is 0.45.

As a result, the *rms* transverse emittance is

$$\varepsilon_{rms} = \frac{1}{4}RP_r /(m_e c) . \tag{7}$$

For the surface state,

$$P_r = \hbar k_{\max}$$

and

$$P_r /(m_e c) = \hbar k_{\max} /(m_e c) = (\hbar c) k_{\max} /(m_e c^2) ,$$

where $\hbar c$ = 197 MeVfm and $m_e c$ = 0.511 MeV. For the Cu(111) surface state, k_{max} = 0.225 Å^{-1} and

$$P_r /(m_e c) = 0.87 \times 10^{-3} .$$

For R = 1 mm, $\varepsilon_{rms} = 0.22 \times 10^{-6}$ m, which is around 1/3 of the value measured in a RF gun [9].

CURRENT STATUS AND FUTURE PLANS

The lab here at LBNL has been steadily built up for the past year and half. In addition to the standard equipment for a surface science lab, we purchased a 2D spatially resolved time-of-flight detector which will greatly speed up data acquisition time and reduce systematic errors generated from moving the sample. At present, all equipment have been set up and the first set of data on Cu(111) has been taken, which shows clearly the dispersion curve of the surface state. Commissioning of both the hardware and the software are well underway.

Once the system is calibrated, we will finish the experiments on Cu, with the hope of shedding some light on the origin of the electrons, especially for a copper cathode in the RF gun. Afterwards, we will focus our attention more and more on developing new photocathode that has the potential of increasing quantum efficiency and decreasing thermal emittance at the same time. One of the more promising candidates is nano-structured cathodes that couples light into the surface through the creation of surface plasmon.

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