

# STUDY OF THE MEAN TRANSVERSE ENERGY AND THE EMISSION MECHANISM OF (N)UNCD PHOTOCATHODES

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## Abstract

Nitrogen incorporated ultrananocrystalline diamond ((N)UNCD) is promising for photocathode applications due to its high quantum efficiency (QE). The mean transverse energy (MTE) which, along with QE, defines the brightness of the emitted electron beam which must be thoroughly characterized and understood for (N)UNCD. Our previous work [APL 114, 093103 (2019)] further corroborated the important role of graphitic grain boundaries (GB's). UNCD consists of diamond ( $sp^3$ -hybridized) grains and graphitic ( $sp^2$ -hybridized) GB's: GB's are behind the high emissivity of (N)UNCD and therefore play a crucial role in defining and controlling the MTE. In this work, the MTE of two different (N)UNCD samples having different ratios of  $sp^3/sp^2$  were measured versus the primary photon energies. As a reference, MTE of highly oriented pyrolytic graphite (HOPG, canonical  $sp^2$ -hybridized graphite) was also measured.

## INTRODUCTION

Challenges in photocathode studies include achieving *i*) high quantum efficiency (QE) *ii*) low thermal emittance/mean transverse energy (MTE) and *iii*) rapid response time. The ratio of the emitted electrons to the beam emittance determines the photocathode brightness. The emitted charge depends directly on the QE of the photocathode material, and the beam emittance is directly related to the MTE of the photoelectrons. The MTE is primarily dependent on cathode material band structures.

(N)UNCD is an *n*-type photocathode that has high electron conductivity through the bulk; it consists  $sp^3$  diamond grains and  $sp^2$  graphitic grain boundaries. Higher QE ( $10^{-3}$ ) [1, 2, 3] may potentially be achieved simply via hydrogen surface termination. The roughness of synthesized (N)UNCD is low ( $<10$  nm) due to its nano-scale crystalline size. Its low physical roughness implies less surface scattering during the process of transporting photoelectrons over the surface barrier. Experimental measurements of the MTE of (N)UNCD as a photocathode was reported by our group [4]. A relatively low average value of 266 meV was demonstrated, while the MTE dependence on the primary photon energy demonstrates a nonconventional behavior. To elaborate on the emission mechanism of (N)UNCD, MTE of two samples with

different  $sp^3/sp^2$  ratios were measured. Additionally, MTE of a HOPG sample was also measured for comparison.

## (N)UNCD SYNTHESIS

The (N)UNCD films were deposited on two highly doped *n*-type silicon substrate by using the microwave assisted plasma chemical vapor deposition (MPCVD) in a  $N_2/CH_4/H_2$  precursor gas mixture. To tune the  $sp^3$ -to- $sp^2$  ratio, the deposition temperature was varied. Raman spectra confirming the physico-chemical UNCD bonding structures of both samples are shown in Fig. 1. The (N)UNCD#1 (red) was grown at 945 °C: it is more graphitized, resistivity measured on intrinsic Si coupon is  $9.5 \times 10^{-5} \Omega \times cm$ . The (N)UNCD#2 (blue) was grown at 860 °C: the film is less conductive with resistivity estimated to be  $2.07 \Omega \times cm$ . The film thicknesses for both samples were found to be  $\sim 430$  nm.

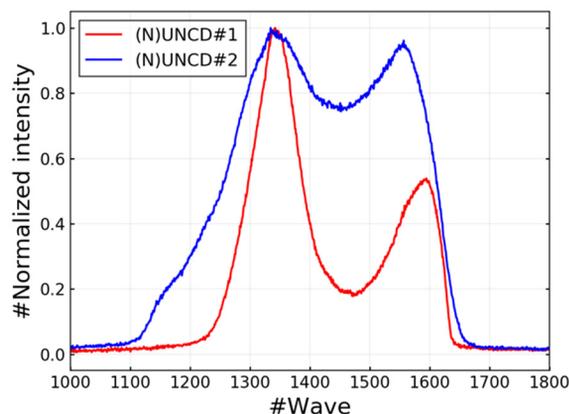


Figure 1: Raman spectra of the two (N)UNCD samples #1 (red) and #2 (blue) showing the characteristic diamond grain ( $\sim 1350$   $cm^{-1}$ ) and graphite GB ( $\sim 1550$   $cm^{-1}$ ) peaks. Laser wavelength was 532 nm.

## EXPERIMENTAL SETUP

The method of the double-solenoid scanning [5] has been used to measure the MTE. The schematic diagram of the experimental setup is shown in Fig. 2.

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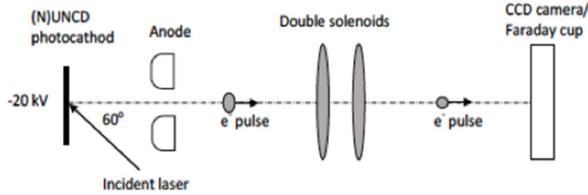


Figure 2: Double-solenoid scan system for MTE measurements, identical to the setup used in Ref.[4].

The electron pulses are generated from (N)UNCD, placed as a photocathode in a 20 kV DC gun, using tunable ultraviolet (UV) radiation from a 30 MHz repetition rate sub-picosecond laser system driven by a diode pumped and mode-locked Yb:KGW laser system [6]. Two solenoids are connected to the same 0-3.0 A constant current power supply with opposite polarities to eliminate the rotation of electrons in the beamline. The CCD camera at the downstream end captures the electron beam profile on the scintillator screen. More information on the experimental setup can be found in Ref.[4].

## EXPERIMENTAL RESULTS

The MTE of metals and many thin film alkali antimonide photocathodes obey the three-step photoemission model extended by Dowell and Schmerge (DS) [7]. In the DS model, the MTE has a linear dependence on the excess photon energy following  $MTE \sim (\hbar\omega - \phi)/3$ , where  $\hbar\omega$  is the primary photon energy and  $\phi$  is the cathode work function.

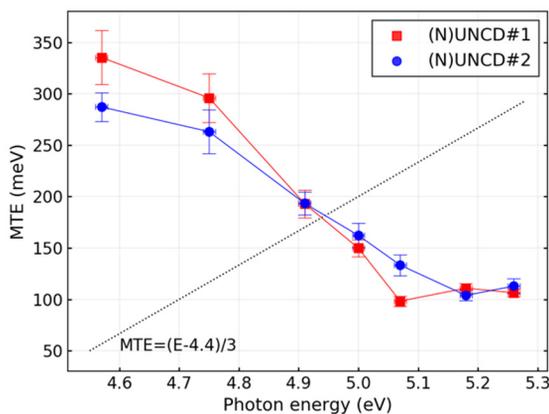


Figure 3: Measured MTE results of two samples labeled as (N)UNCD#1 and (N)UNCD#2 extracted from double-solenoid scan data. The black dotted line is the MTE dependence of metals as per the DS law; the work function of two samples were estimated to be 4.4 eV.

The measured MTE of two (N)UNCD samples are shown in Fig. 3. The two samples have similar MTE behaviors, where the MTE values were found to be decreasing with the increasing photon energies. Additionally, at high photon energies ( $>5.05$  eV), the MTE for both samples tend to remain at a constant level of 100 meV. The black dotted line is the MTE plotted as a function of photon energies following the DS model. Comparisons of the

measured MTE for both (N)UNCD samples to the cathodes that follow the DS model suggest a different emission mechanism.

A possible explanation of this effect is that there are two emission mechanisms involved for the (N)UNCD photocathode. At low photon energies, emission from the sp<sup>2</sup> defect states may dominate, with the photoelectrons likely originating from the GB's. As shown in Fig. 3, the MTE of (N)UNCD#1 is about 50 meV higher than that of the (N)UNCD#2. Then with the increasing photon energies, electrons could begin to be emitted from band states in the diamond nano-crystals. If so, the electrons may originate from the X-valley of the conduction band of diamond. This could explain the low MTE values and the flat MTE trend since the electron effective mass of the X-valley transverse to the (100) crystal direction is small. Due to the conservation of the transverse momentum in the photoemission process, electrons emitted from narrow energy bands (low effective mass) can result in that MTE behavior is less sensitive to the primary photon energy [4], therefore showing trends that are different from the DS law.

The HOPG was studied for comparison and the MTE values are summarized in Fig. 4. In contrast to the two (N)UNCD samples, the MTE of pure sp<sup>2</sup> graphite shows a strong linear dependence on the photon energy in that it obeys the DS law, i.e. it behaves like a classical metal.

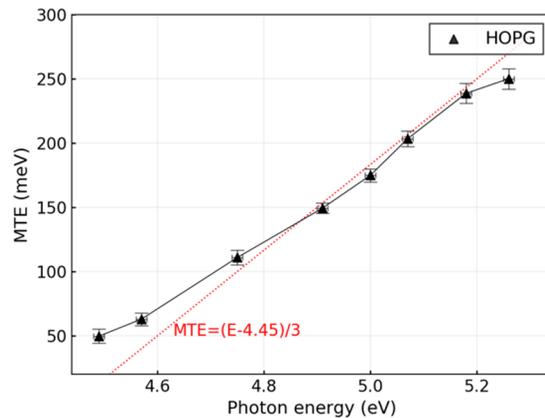


Figure 4: Measured MTE results for HOPG. The red dotted line is the DS model, where the work function of HOPG is taken to be 4.45 eV.

To reveal the detailed emission mechanism of (N)UNCD photocathode, our next step is to measure the quantum efficiency (QE) of both (N)UNCD samples at different photon energies. As stated by T. Vecchione et al [8], the QE of “perfect” metals following the three-step model as

$$QE = S_{12} \left( \frac{Li_2 \left[ -Exp \left[ \frac{e}{kT} (\hbar\omega - \phi) \right] \right]}{Li_2 \left[ -Exp \left[ \frac{e}{kT} \mu \right] \right]} \right) \quad (1)$$

where  $S_{12}$  is a constant and  $\mu$  is the chemical potential. This expression implies a strong correlation between the QE and the excess energies for the metallic cathode. If two distinctly different emission mechanisms are indeed involved for the (N)UNCD, defect state and band emission,

the QE may not follow such power-law dependence. Indeed, one might expect a discontinuity in the QE as a function of excess energy ( $\hbar\omega - \phi$ ) as the postulated band emission starts to dominate over the defect emission.

### CONCLUSION

The MTE of two (N)UNCD samples were measured over a wide range of primary photon energies. MTE values for both samples were found to be decreasing as the excess energies increased. The two emission mechanisms were proposed to explain such unconventional MTE behavior. This could be a great benefit for the photocathode applications. The brightness of (N)UNCD photocathodes could be enhanced by increasing the photon energies because the QE would increase, while the MTE would decrease or remain relatively low and constant.

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