

Transmission photocathodes based on stainless steel mesh and quartz glass coated with N-doped DLC thin films prepared by reactive magnetron sputtering

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Abstract. The influence was investigated of N-doped diamond-like carbon (DLC) films properties on the quantum efficiency of a prepared transmission photocathode. N-doped DLC thin films were deposited on a silicon substrate, a stainless steel mesh and quartz glass (coated with 5 nm thick Cr adhesion film) by reactive magnetron sputtering using a carbon target and gas mixture Ar, 90%N₂+10%H₂. The elements' concentration in the films was determined by RBS and ERD. The quantum efficiency was calculated from the measured laser energy and the measured cathode charge. For the study of the vectorial photoelectric effect, the quartz type photocathode was irradiated by intensive laser pulses to form pin-holes in the DLC film. The quantum efficiency (QE), calculated at a laser energy of 0.4 mJ, rose as the nitrogen concentration in the DLC films was increased and rose dramatically after the micron-size perforation in the quartz type photocathodes.

1. Introduction

Electron emission from carbon materials has been based on two effects: field enhancement from conducting nanostructures and barrier lowering due to the negative electron affinity of surfaces [1]. The photoemission properties of different samples were rationalized by considering the electron emission process located at the a-C/diamond/vacuum triple border and the quantum efficiency governed by the ratio of amorphous sp²-C to crystalline sp³-C [2]. In [3], the authors investigated the electron field emission properties of sulphur-assisted nanocrystalline carbon (n-C:S) thin films grown on molybdenum substrates by the hot-filament CVD technique using a methane-hydrogen (CH₄/H₂) and hydrogen sulphide-hydrogen (H₂S/H₂) gas mixtures. In [4], the results were reported and discussed of the external quantum efficiency (QE) measurements, in the range 150–210 nm, of poly-, nano- and single-crystalline diamond (PCD, NCD and SCD) film photocathodes (PCs). The process of diamond-like carbon deposition by plasma-assisted chemical vapor deposition (PECVD) using deuterated hydrocarbons as precursor gases was investigated in [5]. Diamond-like carbon thin films were deposited from a pure graphite target by DC magnetron sputtering. Experimental

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parameters, i.e., substrate temperature and negative bias voltage, were varied to finely tune the chemical bonding property (sp^2/sp^3) of the as-deposited DLC films [6]. DLC films deposited under a mixture of acetylene/argon plasma over a large area and three dimensional deposition in electron cyclotron resonance (ECR) systems were reported in [7]. The possibility of application of ion implantation for the modification of the physicochemical properties of carbon coatings was presented in [8]. Diamond-like carbon thin films were deposited on p-type Si (100) substrates by the RF hollow-cathode method under different RF power and pressures using ethane as the precursor gas [9]. The photo-gun is a key element of injectors for producing high-charge electron bunches. Research continues on various designs of cathodes. A promising, but technically challenging, DC gun with a hollow cathode was proposed by JINR [10]. In the development of this concept, the use of transmission photocathodes in the form of a grid was the most promising direction. The vectorial photoelectric effect has long been known. It is necessary to notice that research of vectorial photoeffect demands that the photocathode surface be smooth. In the presence of a roughness, the polarized light will have various polarizations in relation to the plane of incidence on variously oriented surface elements of the rough photocathode.

In this work, we investigated the influence of N-doped diamond-like carbon (DLC) films properties on the quantum efficiency of a prepared transmission photocathode. The N-doped DLC films were prepared by reactive magnetron sputtering with Ar as an inert gas and 90%N₂+10%H₂ mixture as a reactive gas. The structural properties of the DLC films were investigated by RBS and ERD measurements. The properties of the prepared transmission photocathodes were studied by measurement of the quantum efficiency. The vectorial photoelectric effect of the quartz type photocathode was studied.

2. Experimental

N-doped DLC thin films were deposited on a silicon substrate, a stainless steel mesh (samples MS), and quartz glass coated with a 5-nm-thick Cr adhesion film (samples MQ), by reactive magnetron sputtering. The mesh grid consisted of stainless steel wire with a diameter of 0.03 mm, the cell size being 0.04 mm. The quartz glass substrates were two-side polished, with dimensions 10×10×0.5 mm. Prior to deposition, standard cleaning was performed to remove impurities from the silicon surface, stainless steel mesh and quartz glass. The magnetron target was a high-purity graphite disk with a diameter of three inches. High-purity argon was used as an inert gas and the gas mixture 90% nitrogen + 10% hydrogen, as a reactive gas. The flow rates of 90%N₂+10%H₂ and Ar gases were 2 sccm and 30 sccm for sample MS2 (MQ2), 4 sccm and 30 sccm for sample MS3 (MQ3), and 8 sccm and 30 sccm for sample MS4 (MQ4). The DLC film for sample MS1(MQ1) was sputtered without a reactive gas. The DLC film was deposited at a working pressure of 0.6 Pa and the magnetron input power was a combination of RF power (200 W at 13.56 MHz) and DC power ($U_{DC} = 550$ V, $I_{DC} = 150$ mA). During the magnetron sputtering, the substrate holder was biased at $U_b = -150$ V. The substrate holder temperature during deposition was 100 °C. The film thickness for the silicon substrate and the stainless steel mesh was about 200-250 nm, and for quartz glass, about 25 nm. The DLC films were deposited on both sides of the mesh under the same deposition conditions. The DLC films on the Si substrate were used for structural characterization. The elements' concentrations in the films were analyzed using Rutherford backscattering spectrometry (RBS) and elastic recoil detection (ERD) simultaneously [11]. A lift-off technique was used for Al mesh preparation on DLC/quartz structures. Fig. 1 shows the technological steps of preparation and shaping of Al mesh on quartz-type transmission photocathodes. For the study of the vectorial photoelectric effect, the quartz-type photocathode was irradiated by intensive laser pulses (three-four pulses with laser power density of about ~5 MW/cm²) to create micron-size perforation in the Cr and DLC films by laser ablation. The transmission photocathode quantum efficiency testing was performed at JINR Dubna. The 15-ns UV laser pulses (quadrupled Nd:YAG laser, 266 nm) with a laser spot size of 5 mm were used to illuminate the backside of the photocathode. To draw the electrons from the DLC-film-coated stainless-steel mesh and the quartz-glass photocathode, the cathode was biased by a negative voltage of roughly 20 kV. The bunch charge was measured by using a Faraday cup (FC) connected to the

ground through a measuring capacitor with a matched cable; the voltage on the charging capacitor (U_c) was monitored by a 500 MHz oscilloscope.

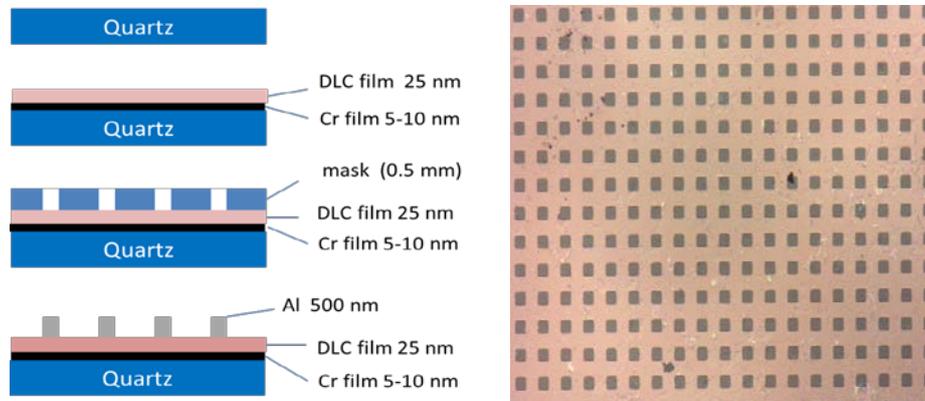


Figure 1. Transmission photocathode technology based on quartz glass coated with a DLC film.

3. Results and discussion

The measured and simulated RBS and ERD spectra for the MS1 to MS4 samples are shown in figure 2. The RBS and ERD analyses indicated that the films contain carbon, nitrogen, hydrogen, and a small amount of oxygen. In figure 2a, the RBS spectra show a leading edge of about ~530 ch corresponding to the Si substrate. The slight difference of the Si edge position between the samples can be attributed to the differences in the films' thickness. The higher counts corresponding to C are observed in the profile of the bulk Si at around ~320 ch. Higher counts corresponding to N are observed in the profile of the bulk Si at around ~370 ch. In figure 2b, the leading edge of H occurs at approximately 530 ch. The concentrations of elements for all samples were: sample MS1(carbon 90 at.%, nitrogen 1-2 at.%, hydrogen 7 at.%, oxygen 1-2 at.%), sample MS2 (70, 15, 14, 1-2), sample MS3 (64, 21, 14, 1-2), and MS4 (61, 23, 15, 1-2), respectively. We assume that the concentration of hydrogen and nitrogen in the MS1 film was mainly caused by the vacuum chamber atmosphere. Adding the gas mixture 90%N₂+10%H₂ to the working gas results in reducing the carbon concentration in the films from 90 at.% to 61 at.%. The hydrogen concentration in samples MS2-4 was practically the same. We also assume that the elements' concentrations in the films for MQ samples were the same as in the MS samples due to the simultaneous preparation in one vacuum cycle. The DLC film sputtering on a quartz substrate takes place at the end of the DLC film sputtering on a stainless steel mesh and silicon to obtain a thickness of about 25 nm. Figure 3a) shows the bunch charge as a function of the laser

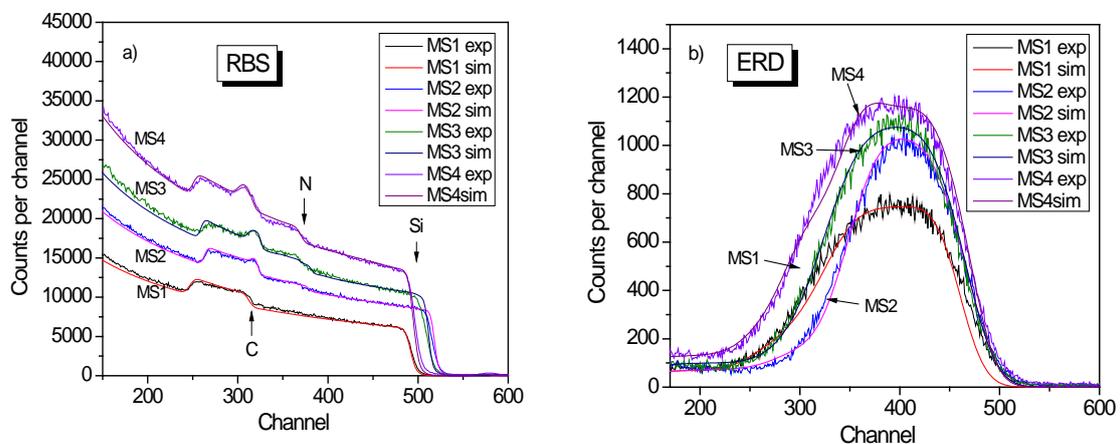


Figure 2. RBS a) and ERD b) experimental and simulated spectroscopy results for samples MS1-4.

energy for MQ prepared photocathodes after micron-sized perforation. The bunch charge as a function of the laser energy for MS prepared photocathodes was about three times lower. The bunch charge increased as the laser energy increased. The increase in the laser energy means an increase in the number of photons causing photoemission, which leads to an increase in the number of photoelectrons produced at the cathode surface. The bunch charge intensity trend of the MQ photocathodes reported in figure 3a evidences a dependence of the photoemission properties on the graphitic component inside the DLC films and the nitrogen concentration. Incorporated nitrogen mainly exists as N-sp²C and N-sp³C bonds and small amounts exist as N-H bonds. The N-N bonds may be related to encapsulated nitrogen [12]. Usually, we measure the extracted charge Q as a function of the laser energy EL . The QE was calculated from a linear fit to the data points at low charge densities, where space charge effects are negligible and the relation $Q(EL)$ is linear. The quantum efficiency was calculated using the formula

$$QE = N_e/N_\nu, \quad (1)$$

where N_e is the number of electrons and N_ν is the number of photons. Figure 3b shows the quantum efficiency as a function of the type of the prepared transmission photocathode. The quantum efficiency for MS type transmission photocathodes were: MS1 – 0.9×10^{-4} %, MS2 – 1.0×10^{-4} %, MS3 – 1.5×10^{-4} % and MS4 – 1.8×10^{-4} %. The quantum efficiency for MQ type transmission photocathodes after micron-size perforation were: MQ1 – 4.4×10^{-4} %, MQ2 – 5.0×10^{-4} %, MQ3 – 7.2×10^{-4} % and MQ4 – 1.8×10^{-4} %. On the basis of the results presented, it can be concluded that the QE of the MQ photocathode is higher than the QE of the MS photocathode. The emission mechanism in DLCs has been difficult to understand. The experimental evidence suggests that the main barrier to emission from a DLC is large and at the front surface. Lowering the barrier requires some types of heterogeneity [13]. In our case, the results obtained show that lowering the barrier can be achieved by nitrogen doping of the DLC film. The quantum efficiency (QE) calculated at a laser energy of 0.4 mJ rose as the nitrogen concentration in the DLC films was increased, and dramatically rose after micron-size perforation of the quartz type photocathodes. The vectorial photoelectric effect was observed in all MQ and MS cathodes investigated, but was much more efficient in the MQ cathodes. In the case of MS cathodes, the effect is suppressed, as the net-substrate geometry does not allow sending p polarized laser light at an optimal angle, so that the highest possible amount of charge for a given laser energy cannot be achieved. These results are not only useful in the optimization of a photocathode, but also allow a deeper understanding of the photoemission physics.

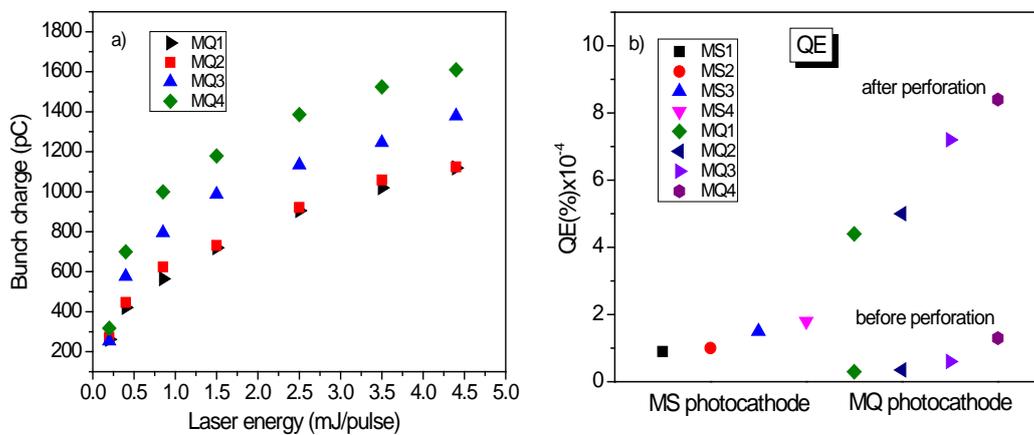


Figure 3. Bunch charge versus laser energy (a) for MQ transmission photocathodes and quantum efficiencies (b) for MS photocathodes and MQ photocathodes before and after micron-size perforation.

4. Conclusions

We investigated the influence of N-doped diamond-like carbon (DLC) films properties on the quantum efficiency of prepared transmission photocathode. N-doped DLC thin films were deposited on a silicon substrate, a stainless steel mesh and quartz glass (coated with a 5-nm-thick Cr adhesion film) by reactive magnetron sputtering. The RBS and ERD analyses showed that the films contain carbon, nitrogen, hydrogen and a small amount of oxygen. The nitrogen concentration rose with the higher flow of reactive gas and reduced the carbon concentration. A microelectronic technology was used for preparation of quartz-glass-type transmission photocathodes. The quantum efficiency (QE) calculated at a laser energy of 0.4 mJ rose with the increase in the nitrogen concentration in the DLC films for all transmission photocathodes and dramatically rose after micron-size perforation of the quartz-type photocathodes. The best results for the QE (%) was 8.4×10^{-4} for a quartz-type transmission photocathode. The results indicate that the surface properties are critical for the vectorial photoelectric effect.

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References

- [1] Nemanich R J, Bilbro G L, Bryan E N, Koeck F A, Smith J R and Tang Y 2010 *Proc. 3rd Int. Conf. Nanoelectronics (IEEE010)* ed Paul K Chu (2010 Hong Kong) p 56
- [2] Sessa V, Orlanducci S, Fiori A, Terranova M L, Tazzioli F, Vicario C, Boscolo I, Cialdi S and Rossi M 2005 *Proc. SPIE* **5838** p 216
- [3] Gupta S, Weiner B R, Weiss B L and Morell G 2001 *Proc. Symp Mater. Res. Soc.* (2001 San Francisco) **675** W6.9.1
- [4] Nitti M A, Colasuonno M, Nappi E, Valentini A, Fanizza E, Bénédic F, Cicala G, Milani E and Prestopino G 2008 *Nucl. Instrum. Methods A* **595** 131
- [5] Maia da Costa M E H and Freire Jr F L 2010 *Surf. Coat. Technol.* **204** 1993
- [6] Lu Z G and Chung C Y 2008 *Diamond & Related Materials* **17** 1871
- [7] Guo C T and Ueng H Y 2014 *Vacuum* **107** 304
- [8] Batory D, Gorzedowski J, Rajchel B, Szymanski W and Kolodziejczyk L 2014 *Vacuum* **110** 78
- [9] Pang X, Peng H, Yang H, Gao, Wu X and Volinsky A A 2013 *Thin Solid Films* **534** 226
- [10] Dudarev A, Budagov U A, Balalykin N, Kobets V, Lyablin M V, Sabirov B M, Shirkov G, Syresin E and Trubnikov G V 2013 *Proc. IPAC 2013* (2013 Shanghai) p 1631
- [11] Kobzev A P, Huran J, Maczka D and Turek M 2009 *Vacuum* **83** S124
- [12] Liu Y, Yu H, Quan X, Chen S, Zhao H and Zhang Y 2014 *Sci. Rep.* **4** 6843
- [13] Robertson J 2003 *IEICE Transaction on Electronics* **E86-C (5)** 787