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Secondary electron emission properties of III-nitride/ZnO coaxial heterostructures under ion and X-ray bombardment

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Abstract

The secondary electron emission (SEE) yield of heterostructures of zinc oxide (ZnO) nanoneedles coaxially coated with aluminum nitride (AlN) or gallium nitride (GaN) has been studied using ion and X-ray beams. This paper describes experiments performed with ions (2 MeV protons and 3.6 MeV/nucleon carbon beams) and photons (synchrotron radiation at ≈1 keV). The SEE yield of the heterostructures is enhanced significantly by the intrinsic nanostructure of the ZnO nanoneedle templates as compared to the AlN and GaN thin films on silicon (Si) substrates [T.J. Vink, R.G.F.A. Verbeek, V. Elsbergen, P.K. Bachmann, Appl. Phys. Lett. 83 (2003) 2285]. One of the mechanisms responsible for SEE yield enhancement can be attributed to the larger area of the nanostructured surface.
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1. Introduction

Much work has been done with these new one-dimensional (1D) nanostructure materials because of their excellent field emission properties [1–9]. There is a high potential for developing universal detectors based on these nanomaterials. These detectors will offer higher sensitivity than existing ones because of the higher efficiency in producing secondary electrons. A similar idea was previously developed for carbon foils [10], boron-doped diamond [11] and is now proposed for nanomaterials [4]. We have performed a series of experiments with ions and X-rays to better understand processes governing the SEE in 1D nanostructured materials. Similar work has been published for SEE emission [12–14] and field emission [15–18]. It is found that the SEE yields are significantly enhanced by the intrinsic nanostructure of the ZnO nanoneedle template. The results also help to understand the electron emission characteristics of 1D nanostructures.

2. Experiment

ZnO nanoneedles were grown by catalyst-free MOlecular Chemical Vapour Deposition (MOCVD) on Si substrates using diethylzinc (DEZn) and oxygen gas in the 400–500 °C range [19]. Following the fabrication of ZnO

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nanoneedles, around 10 nm thick AlN or GaN layers were deposited by low pressure metal-organic vapor phase epitaxy (MOVPE) directly on the ZnO nanoneedles using trimethyl-Ga (TMGa) and trimethyl-Al (TMAl) and ammonia (NH_3) as precursors, respectively. Details of the growth conditions and structural characterizations of the coaxial heterostructures have been reported elsewhere [20]. The SEE properties of ZnO-coated carbon nanotubes (CNTs) using biasing technique in a scanning electron microscope [12] has been investigated. During our studies we performed analysis of the SEE properties for several materials including Au, Boron-doped CVD diamond, silicon, silicon with coating (e.g. ZnO, GaN, AlN, MgO), carbon nanotubes (CNT), carbon nanotubes with coating (e.g. ZnO/CNT, MgO/CNT, AlN/CNT, GaN/CNT), ZnO nanorods, ZnO nanorods with coating (e.g. AlN/ZnO, GaN/ZnO) using ions and X-rays.

Details of the experimental set-up for electrons and ions is presented in Fig. 1. Similar set-up has been used for X-rays. In order to characterize the SEE for ions we have applied the 3.6 MeV/nucleon carbon beam at the heavy ion microbeam facility at GSI, Darmstadt, Germany. In this case the beam was focused to about 1 μm diameter and scanned over $100 \times 100\ \mu m^2$ area. We measured the shape of the secondary electron yield by detecting the signal of the channeltron [21]. We performed experiments on selected samples. Results are presented for GaN/ZnO (Fig. 2(a)) and CsI (Fig. 2(b)). It is clearly visible that the spectrum for the GaN/ZnO heterostructure extends up to about channel 3000 (with amplifier gain 10×) indicating a higher SEE yield as compared to CsI which finishes at about channel 1500 (with amplifier gain 100×). However, the fact that the spectrum for GaN/ZnO extends also to lower channels indicates that the SEE yield from the sample has not been very uniform on the nanoscale level. For comparison, the data for CsI form a characteristic peak with Gaussian distribution which indicates that the sample is more uniform in production of the SEE yield. The CsI sample is selected for presentation as it exhibits the SEE yield higher than Au. For the uniform sample the narrow peak will be formed in the spectrum at the higher channel number with the position of the peak proportional to the SEE yield. It should be noted that the channeltron detector at 2.4 kV voltage is not strictly linear because its gain tends to saturate at high electron current. This means that the improvement in the SEE yield from GaN/ZnO to CsI may be greater than it appears from Fig. 2. The channel number (horizontal axis) is related to the SEE yield in a non-linear way and this dependence has not been established for the system at GSI. The vertical axis corresponds to the number of counts registered by the channeltron detector.

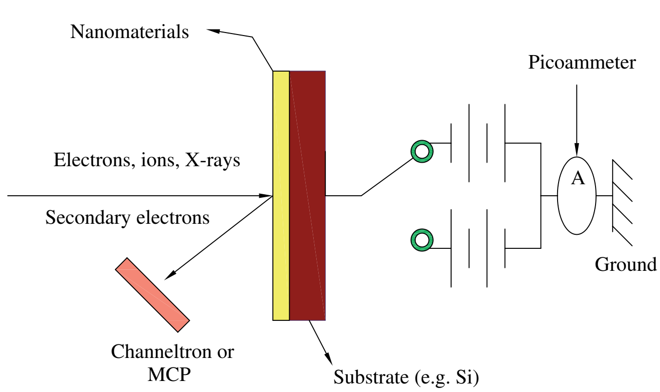

Fig. 1. Schematic diagram of the experimental set-up and new detector with nanomaterials. Secondary electrons generated by the electrons, ions and X-rays are detected with an electron multiplier (channeltron) or a micro-channel-plate (MCP) detector connected with the proper electronics [21]. SEE yield with electrons was characterized by measuring of the current [22]. The channeltron could offer the rising time of the signal down to 2 ns while the MCP down to 250 ps respectively which will allow to develop a very fast detection system assuming a fast release of secondary electrons from the sample.

To investigate the SEE yield and uniformity for different samples we used the high resolution nuclear microprobe at the centre for ion beam applications (CIBA) at the National University of Singapore (NUS). The beam of 2 MeV protons has been focused to about $150 \times 200\ nm^2$ and scanned over ca. $5 \times 5\ \mu m^2$. The secondary electrons have been detected using AMPTEK (MD 502) detector. On average the GaN/ZnO sample showed much better SEE yield than other samples which is consistent with measurement performed with heavy ions at GSI. Fig. 3 shows the result for SEE yield from the sample of ZnO nanorod sample coated with GaN (GaN/ZnO). The lighter color (white) indicates a higher SEE yield and the darker (black) a lower one. Similar data has been obtained using images collected with the SEM at GSI. Thus, the SEE yield of these 1D nanomaterials under proton or electron bombardment is not uniform across the surface.

In order to investigate the SEE emission for X-rays we used the SINS (surface, interface and nanostructure science) beamline at the SSLS in Singapore, details of which can be found in [22]. We performed X-ray photoemission spectroscopy (XPS) using a beam of 1 keV X-rays. The results of the SEE yield are shown in Fig. 4 as a function of the electron energy. It is clearly visible that GaN/ZnO exhibits the highest SEE yields.

3. Results and discussion

After careful analysis of data for many different materials under irradiation of ions and X-rays we found that AlN/ZnO and GaN/ZnO produced much better SEE yield than other materials investigated. These proposed new nanomaterials are highly efficient emitters of secondary electrons and are much better than diamond or metals (e.g. gold) which have been routinely used in the past as efficient SEE. In conclusion, the AlN/ZnO and GaN/ZnO coaxial heterostructures have a higher SEE yield than AlN and GaN deposited on Si substrates. The SEE yield of

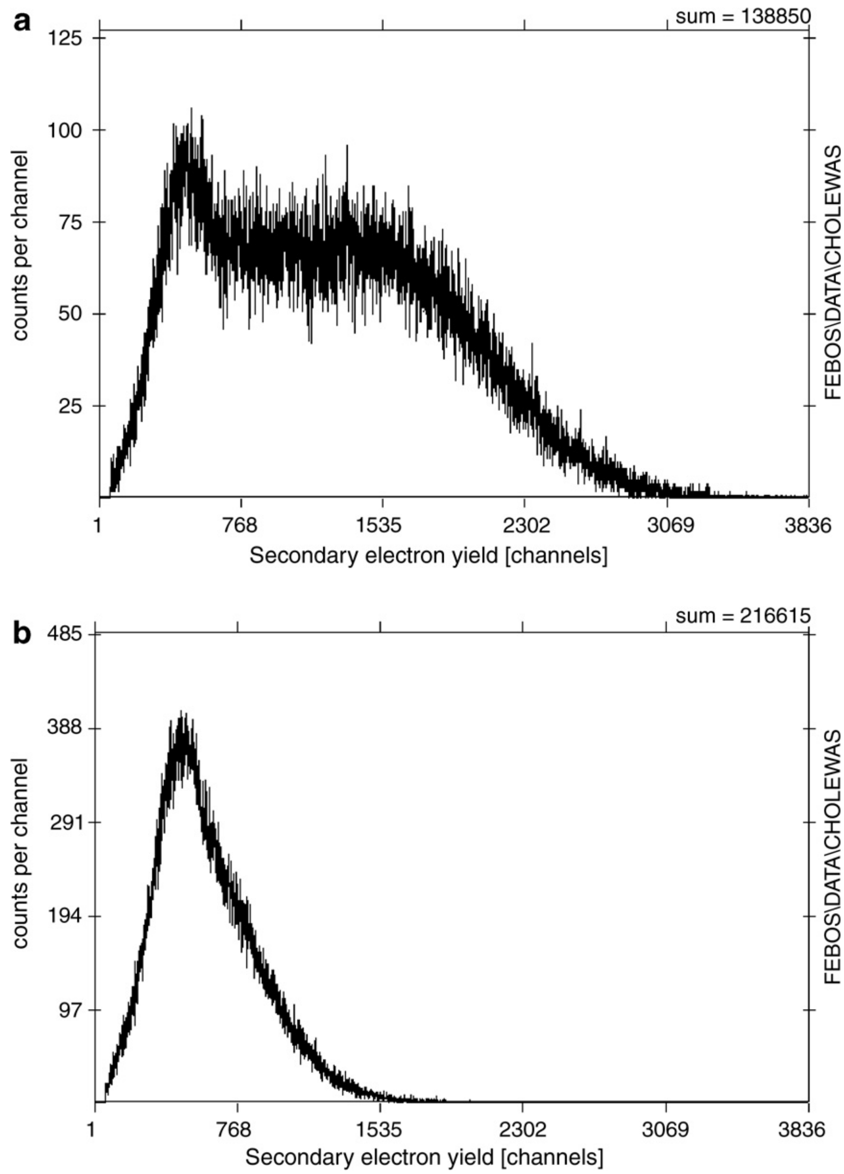

Fig. 2. Secondary electron spectra for GaN/ZnO (a) and CsI (b) measured with a channeltron detector. Bombarding ions: 3.6 MeV/nucleon carbon. Both spectra have been measured under identical conditions using +2.4 kV operating voltage for the channeltron plus an attracting bias of +180 V. The horizontal axis corresponds in a nonlinear way to the SEE yield from the sample. The vertical axis shows the number of counts registered by the channeltron. Data for GaN/ZnO sample were collected with amplifier amplification 10×, while for CsI with amplifier amplification 100×.

the heterostructures is enhanced significantly by the inherited nanostructures from the ZnO nanoneedle templates. These results suggest that the enhancement of SEE in the coaxial heterostructure is due to the combined effect of III-nitride layer and nanostructure. However, the understanding of the mechanisms that contribute to these excellent properties is still limited.

As presented discussed by Fischer et al. [21] the secondary electrons generated from the sample are collected either with a channeltron or an MCP. As a channeltron and an MCP offer an output signal rise time of down to 2 ns and 250 ps, respectively, detectors may be made very fast, provided, the act of secondary electron emission from the sample is also fast enough. Such detectors could be used in fast time-of-flight systems where time resolution below 100 ps could be achieved by using only a small fraction of the output signal. Moreover, the 1D nanomaterials reported are even more promising than boron-doped CVD diamond [11,23] and hence would enable detectors to be produced with improved efficiency. However, several issues including uniformity of the surface still need to be addressed. While most of materials used in radiation detectors are good only for detection of one type of radiation (e.g. ions, or electrons or X-rays) these new nanomaterials seems to have higher SEE efficiency for electrons [12] ions and X-rays used in our research. This property alone could make them extremely useful for developing universal radiation detectors.

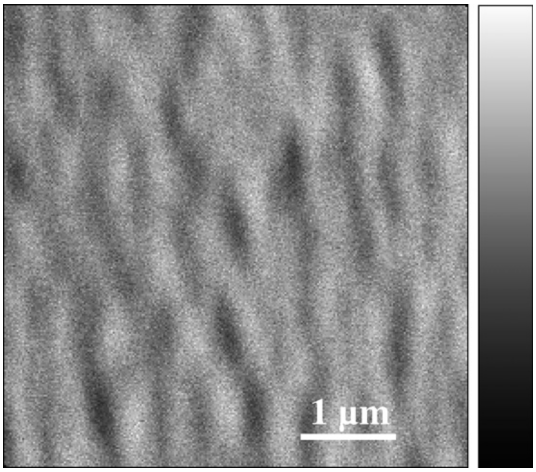

Fig. 3. A map of secondary electron yield from GaN/ZnO measured with a channeltron detector and 2 MeV protons. The scan is $5 \times 5\ \mu m^2$ and the beam focused to ca. $150 \times 200\ nm^2$. The lighter colour (white) indicates higher and the darker (black) lower SEE yield.

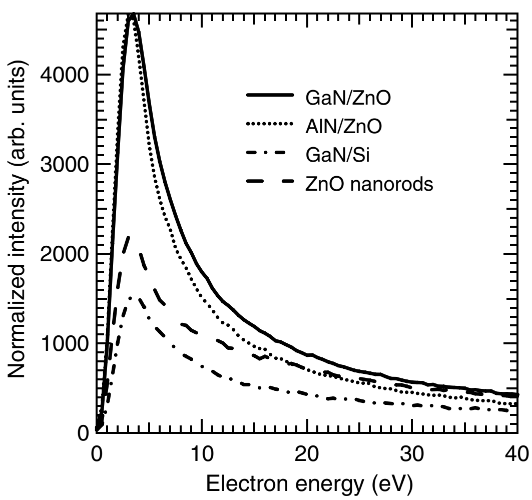

Fig. 4. XPS spectra of four different samples at 1 keV photon energy. The energy of the electrons was measured between 0 and 40 eV. The data for different samples were normalized to the electron beam current of the storage ring, i.e., to the incident photon flux. No bias on the sample.

Acknowledgements

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