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NEGATIVE ELECTRON AFFINITY GALLIUM ARSENIDE PHOTOCATHODES BASED ON OPTICALLY RESONANT NANOSTRUCTURE*

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Abstract

We report the design and fabrication of a new type of negative electron affinity (NEA) gallium arsenide (GaAs) photocathode with optically resonant nanostructures. We observed a significant enhancement of the quantum efficiency (QE) from the GaAs photocathode with nanowire arrays (NWA) due to the Mie resonance effect within the intended wavelength range. Theoretical calculations of the expected reflectance behaviour together with experimental results of optical and photoemission characteristics are presented.

INTRODUCTION

For many years, NEA GaAs photocathodes have been successfully used in electron sources for electron accelerators and advanced light sources [1]. However, ever more challenging facilities are being proposed that require higher quantum efficiency (QE), longer operating lifetime, improved brightness and sometimes requiring polarized beams. Photoemission structures, such as graded doping/band-gap [2,3], strained superlattice [4], and distributed Bragg reflector [5] structures, have been explored to increase the QE or spin polarization of GaAs-based photocathodes. However, all of these photoemission materials rely on GaAs-based epitaxial thin films as the active layer of the photocathodes. The QE of these thin film photocathodes is adversely affected by the low absorption of the incident light. The reflectivity over the wavelength region of interest for GaAs photocathodes with planar geometry is around 30%~40%, and increasing laser power is both costly and can damage the photocathode through heating. In addition, the photoelectrons can only be emitted when they have been transported to the cathode surface, which increases the chance of recombination of photoelectrons and decreases QE [6].

The optically localized Mie resonance (LMR) effect of semiconductor nanostructures has been confirmed in photodetectors, solar cells, and lasers [7,8]. By tailoring the LMR properties, through control of the physical dimensions of the nanostructures, one can significantly enhance light absorption and localize the optical and electric field intensity. Semiconductor nanostructure geometry is a promising alternative to planar types because it offers enhancement of optical absorption by confining, coupling and trapping incident light through the Mie resonance effect, and possible shortening electron transport distance to the surface by localized electric field before recombination can occur. We have fabricated and characterized NEA GaAs nanowire array (NWA) photocathodes and successfully confirmed the Mie resonance effect, by observing a reduction in reflectivity from the GaAs NWA and an increase in photocathode QE, within the design resonant wavelength range.

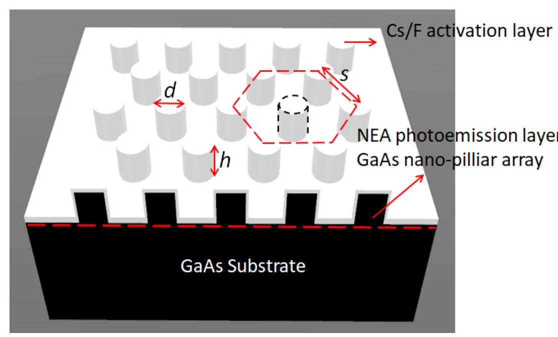

Figure 1: Illustration of structure of a NEA GaAs photocathode device.

CATHODE DESIGN AND EXPERIMENTAL SETUP

Several GaAs photocathodes with two-dimensional periodic NWA structures were designed, fabricated and experimentally studied. Figure 1 shows the illustration of the structure of such GaAs NEA photocathode devices. The GaAs substrate was Zn doped with concentration approximately $(1\sim3)\times10^{19}\text{cm}^{-3}$. The hexagonally distributed GaAs resonant NWA structure was directly fabricated on the GaAs substrate, which is the active photoemission region of the NEA photocathode. Previous reports of high refractive index semiconductor nano-resonators show optical resonance characteristics are mainly affected by the dimensional parameters of the nano-features and optical constants. In this work, the optical constant of the GaAs was specified within the design wavelength range, and the NWA dimensional parameters diameter (d), height (h) and spacing (s) were adjusted to meet the appropriate optical resonance condition for a desired laser

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wavelength. Cesium (Cs) and nitrogen trifluoride (NF_3) were used to for photoemission activation.

The hexagonal GaAs resonant NWA structure was directly fabricated on the GaAs (100) substrate through substrate-conformal imprint lithography (SCIL), per the following process. A 200-nm-thick silicon dioxide (SiO_2) layer was first deposited onto the GaAs (100) substrate using plasma-enhanced chemical vapour deposition (PECVD). A nanowire array master pattern was then fabricated on a silicon (Si) wafer by electron-beam lithography and reactive ion etching. Then a poly-di-methyl-siloxane (PDMS) nano-hole array stamp was used to duplicate the pattern of the Si master wafer. The stamp was composed of two layers of PDMS: a high-Young's modulus layer (H-PDMS) which maintains the nano features and a flexible low-Young's modulus layer (L-PDMS) which allows conformal contact to the substrate.

The flexible stamp can then be applied - using a dedicated SCIL imprint mechanical tool - onto a polymethyl methacrylate (PMMA) photoresist layer that was spin-coated onto the SiO_2 layer of the GaAs substrate. The sample was then subjected to reactive ion etching (Tegal 903e) to transfer the pattern to the SiO_2 hard mask, exposing the etching window. Finally, an optimized inductively coupled plasma etch (Oxford Plasmalab System 100 ICP180) was carried out to obtain the GaAs NWA. The PMMA was removed with acetone, and the SiO_2 mask layer was then removed with buffered oxide etchant, leaving the NWA structure successfully fabricated on the GaAs substrate.

The dimensional parameters of the NWAs depend on the SCIL master pattern and the ICP etching time. The morphology of the GaAs NWA was imaged with a field-emission scanning electron microscope (SEM, NOVA NANOSEM 450). Reflectance spectra were measured using a spectrometer (NOVA-EX, Ideaoptics Instruments, China) at an incident angle of 0°.

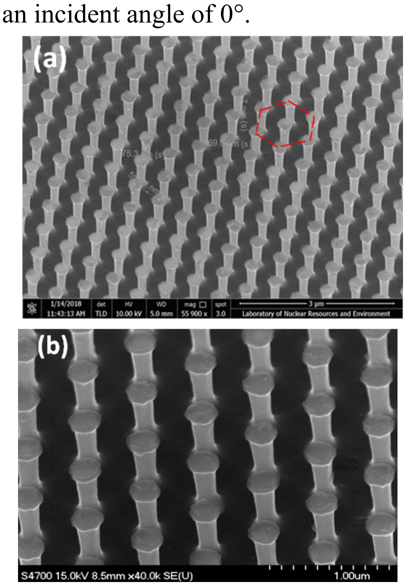

Figure 2: SEM images of the regular hexagonal GaAs NWA fabricated by SCIL: (a) Before heat clean or activation; (b) After heat clean or activation.

Figure 2 shows the SEM image of the GaAs NWA fabricated by SCIL, which indicates that the regular hexagonal GaAs NWA structures were well aligned and vertical. Figure 2b indicates that the original pattern of the nanowire array remained unchanged, with no deformation or damage after multiple cycles of heat cleaning to 550°C and chemical activation.

To obtain the required negative electron affinity (NEA) state for the GaAs NWA structure photocathode, it must be installed in an ultra-high vacuum chamber (base pressure ~10^{-11}Torr) and chemically activated. After baking the chamber at 200°C to remove water vapour, the sample was heated up to 550°C to remove oxides and other contaminants, cooled to room temperature and activated to achieve a negative electron affinity condition using the standard yo-yo activation procedure with cesium and NF_3. A broadly tunable super-continuum light source (NKT Photonics) provides milli-Watts of well-collimated light from 400 to 850 nm for QE measurement of GaAs NWA photocathodes.

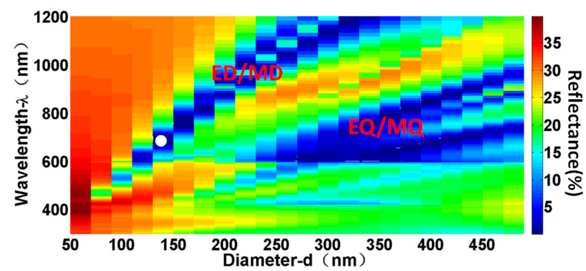

Figure 3: Predicted surface reflectance spectra for GaAs NWA structures as a function of nanowire diameter (d), where height (h) and spacing (s) were set to 500nm and 600nm, respectively.

SIMULATIONS

A number of NWA structures were fabricated to determine the effect of the dimensional parameters diameter (d), height (h) and spacing (s) on the Mie resonance. Prior to fabrication, finite difference time domain (FDTD) electromagnetic simulations were performed to predict absorption dependence on wavelength. Figure 3 shows a simulation of surface reflectance spectra as a function of d for the GaAs NWA structures on GaAs substrate, where h and s were set to 500 nm and 600 nm, respectively. It can be seen that the electric and magnetic dipole (ED/MD) and quadrupole (EQ/MQ) mode resonances can be excited by varying the fabrication parameters. For the typical 600-800 nm wavelength range associated with GaAs photocathode applications the ED/MD mode resonances were excited for the diameter range of ~ 110-170 nm and the EQ/MQ mode resonances were excited diameter range ~ 240-380 nm.

Figure 4 shows a simulation of the electric field intensity ($|E|^2$, colour bar scale) for GaAs nanowire diameter d=140 nm on a GaAs substrate, for the dipole resonance wavelength λ = 727 nm, shown by the white dot in Figure 3. The vertical direction of the image is parallel to the E-field of the linearly polarized incident light. The $|E|^2$ peak

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along the vertical walls of the NWA structure indicates that under resonance conditions, most of the incident light will be absorbed in the GaAs nanowires, which should lead to increased QE and decreased reflectivity.

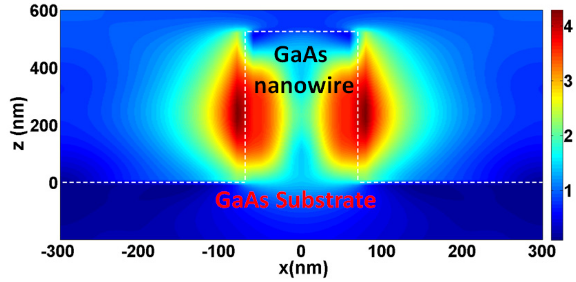

Figure 4: A plot of the electric field intensity (|E|2, colour scale) as a function of position along the photocathode surface, showing the simulated dipole resonance mode profile within a vertical cross section of the GaAs nanowire resonator on the GaAs substrate (d = 140 nm and λ = 727 nm) in the plane parallel to the E-field of the linearly polarized incident light.

RESULTS AND DISCUSSION

Experimental surface reflectance spectra are presented in Figure 5, for GaAs photocathodes with and without the NWA structure, together with theoretical predictions. The NWA sample (d = 140 nm, h = 525 nm and s = 600 nm) shows a marked decrease in reflectivity within the wavelength range 700-800nm compared to the non-patterned film, corresponding to the calculated dipole resonance for these dimensions. The shape of the GaAs nanowires investigated was cylindrical in the theoretical simulation, but the shapes of the experimentally fabricated nanowire arrays are not exactly cylindrical (See in Figure 2). We believe this explains the difference measured and predicted reflectance spectra in Figure 5, especially at short wavelengths (500-60nm).

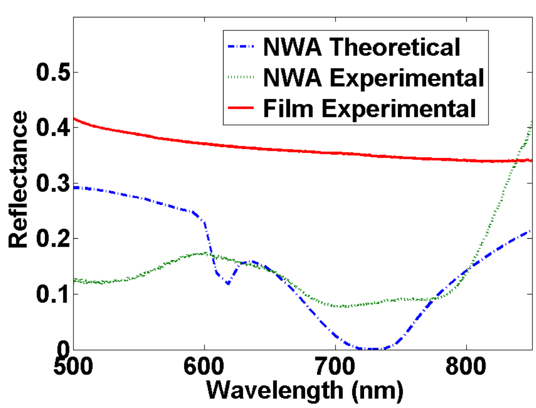

Figure 5: Measured and predicted surface reflectance spectra for GaAs photocathode with and without the NWA resonator (the dimensional parameters: d = 140 nm, h=525nm and s=600nm).

Figure 6 compares the calculated absorption spectrum and the experimental QE of both a non-patterned GaAs film and the GaAs NWA structure resonator, again with d=140 nm, h=525nm and s=600nm. The increase in QE for the NWA structure is evident within the wavelength resonance region between 700-800 nm. These experimental results provide solid evidence that nano-structure technology is indeed an effective method to enhance the QE of GaAs photocathodes, and that QE can be increased within the wavelength range required for high polarization photoemission sources.

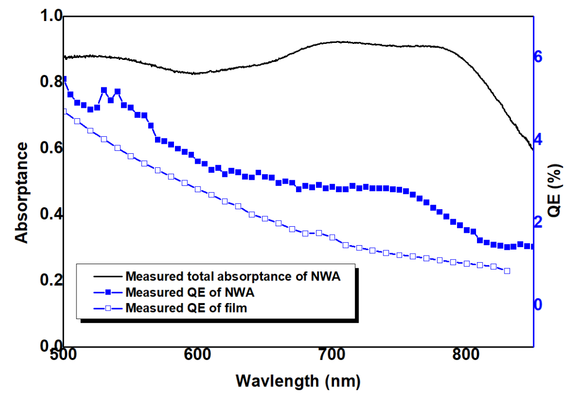

Figure 6: Measured light absorption together with QE for GaAs photocathodes with and without the NWA resonator structure.

SUMMARY

We described GaAs nanowire resonance structures grown on GaAs substrates that served to improve the QE of NEA GaAs photocathodes. FDTD simulation results show that by varying geometric parameters of the NWAs, both dipole and quadrupole resonance modes could be excited in GaAs NWA for the critical wavelength range of 700-800 nm required for polarized photoemission from GaAs. Under the resonance condition, the total surface reflectance decreased to 5% compared to typical 30 – 40%, and most of the incident light could be confined and absorbed in GaAs nanowire active region. Now that QE enhancement has been shown using a NWA structure on GaAs, other critical beam parameters such as polarization, emittance and operational lifetime will be measured in future studies to qualify the NWA structure for use as a high polarization photoelectron source. The QE obtained in this work was lower than what would be from an NEA-GaAs photocathode (~20%), which may be a result of surface damage introduced in the GaAs nanowire etching process. We work to manufacture better samples in a near future work. It should be noted that the goal of present work was to demonstrate the expected physical effect, instead of achieving the highest possible QE.

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