

NANOSTRUCTURED PHOTOCATHODES FOR SPIN-POLARIZED ELECTRON BEAMS*

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Abstract

We present progress on incorporation of nanopillar arrays into spin-polarized gallium arsenide photocathodes in pursuit of record high tolerance to ion back-bombardment. Our goal is to exceed the 400 Coulomb record for a high polarization milliampere-class electron source set at Jefferson Laboratory in 2017, while maintaining high quantum efficiency (QE) and spin polarization with a superlattice.

Because the Mie effect is resonant, uniformity and careful control over nanostructure geometry is key. We report excellent uniformity and straight sidewall geometry with improved optical absorption using a painstakingly optimized inductively coupled plasma reactive ion etch. We also report the application of Kerker theory to spin-polarized photocathode nanopillar arrays, setting new requirements on nanostructure dimensions to avoid spoiling spin polarization. Finally, we also report initial steps toward re-establishing U.S. production of strained superlattice photocathodes towards integration with nanopillar arrays.

INTRODUCTION

Nanopillar arrays (NPAs) in gallium arsenide (GaAs) photocathodes have been reported to enhance quantum efficiency (QE) [1-2]. Spin polarization (P) and charge lifetime in the presence of ion back-bombardment was not quantified. These prior works also used p-type (zinc-doped) bulk GaAs, rather than state-of-the-art superlattices.

We have extended prior work in the present study to optimize the NPA nanofabrication process, to be discussed in the Fabrication section. We are preparing to measure cathode lifetime and anticipate it may increase significantly: non-normal nanopillar surfaces receive low ion bombardment but may contribute most of the QE with appropriate tuning of the dipole Mie resonance via NPA dimensions. Optical Mie resonance in nanopillar arrays preferentially traps light near nanopillar sidewalls. In a direct current gun, nearly zero back-bombardment ion flux will reach vertical sidewalls. The portion of QE attributable to sidewall emission is anticipated to tolerate back-bombardment.

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KERKER THEORY

Light in Mie resonance can be depolarized resulting in deterioration of circular polarization. GaAs requires pure circular polarization to achieve high spin polarization of photoelectrons. Special conditions of conserving polarization are known as Kerker's conditions [3]. The polarization is preserved if the magnetic and electric polarizabilities of the scatterer are equal, resulting in suppression of backward scattering. We have performed simulations of Mie scattering with the miepython package (Fig. 1), where we plot the absorption efficiency of a GaAs sphere (orange curve) and the backscattering efficiency (blue curve) versus the size parameter, defined as $x = \pi d/\lambda$ where d is the sphere diameter and λ the optical wavelength.

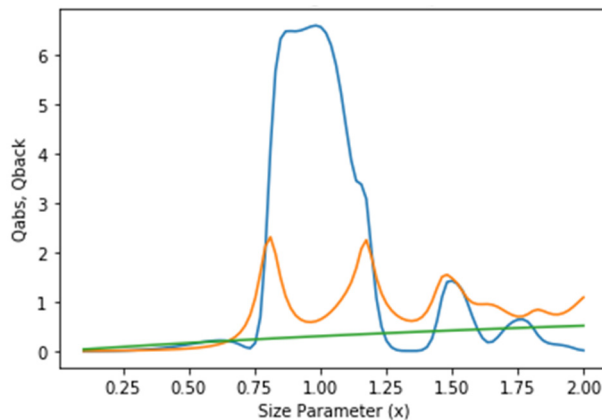


Figure 1: GaAs sphere Mie efficiency for backscatter (blue, Q_{back}) and absorption (orange, Q_{abs}), vs. absorption of infinite slab, thickness equal to sphere diameter (green).

Based on the simulation, we can optimize a choice of nanostructure dimensions to simultaneously enhance QE and preserve photon polarization, hence preserve spin polarization of the photoemitted electron beam. We note this has been done successfully in non-photoemissive studies: the zero backscattering condition has been experimentally demonstrated in single GaAs nanoparticles [4].

Kerker's conditions lead to a non-intuitive design paradigm for Mie-resonant structures in spin-polarized photocathodes. Instead of optimizing on resonant absorption for QE, minima should be found in the back-scattering curve (in Fig. 1, at $x=0.75$ and $x=1.3$ to 1.4) which restricts the nanopillar size parameter, and then the best absorption for QE found in this subset (in Fig. 1, at $x=0.75$). Work is progressing to do this for cylindrical structures on planar substrates and for nanopillars capped by superlattices.

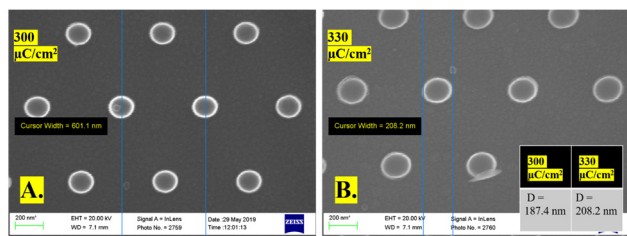


Figure 2: SEM of resist diameter vs dose after developer.

NANOPILLAR ARRAYS

Fabrication and Optimization

We have fabricated nanopillar arrays in GaAs 100 with pitch $s = 600$ nm, diameter $d = 290$ nm, and height $h = 350$ nm, using an optimized e-beam lithography write on a Raith 150 e-beam lithography machine at the Argonne National Laboratory's Center for Nanoscale Materials. Previous work used substrate-conformal imprint lithography (SCIL), and in SCIL multiple process steps make optimization difficult: e-beam write and etch of a master silicon wafer, transfer of the pattern to a polymer, and multiple etch steps. The resulting NPA sidewalls were very concave [1].

To improve upon these results, we opted for a minimum of process steps: a negative resist spin coat (which later serves as the etch mask), direct e-beam write on the resist-coated GaAs wafer, development and descum, and inductively coupled plasma (ICP) reactive ion etch (RIE). Spin-coating must be thin enough for the e-beam to fully expose throughout the resist; for our MaN 2405 resist we use 355 nm thickness. In Fig. 2, diameter is highly sensitive to dose during e-beam write; to achieve 190 nm diameter we use 300 $\mu\text{C}/\text{cm}^2$ at 30 kV. Development and descum must be kept short to avoid undercutting of the negative resist, leading to tapered sidewalls after the etch. The ICP RIE (Oxford PlasmaLab 100) is optimized for pressure, gas flow rates for Ar, Cl_2 and BCl_3 , ICP power and RF power. After ~ 20 etch iterations, we optimized for excellent sidewall straightness. The final recipe was 2 sccm Cl_2 and BCl_3 , 10 sccm Ar, 5 mTorr chamber pressure, 50 W RF, and 400 W ICP power at 20°C.

Characterization

Figure 3 shows the NPA after etching and resist removal, and we observe significant improvement in nanopillar geometry compared to the SCIL process [1]. Spectrophotometric reflectance is excellent compared to planar GaAs (Fig. 4). The reduction in reflectance is improved by the straight sidewalls and uniform geometry; by comparison the SCIL process with concave sidewalls was limited to a 10% specular reflectance minima. QE in our NPA should increase relative to that result.

Superlattice Progress and Future Work

We have fabricated a 20-layer InGaAs-AlGaAs superlattice cathode at the MBE facility at University of Arkansas, using a dedicated III-V growth chamber, following the SLSA 2 recipe from Nishitani et al [5]. This superlattice

cathode has been delivered to Euclid and is now being etched for NPA. A new etch optimization is required with new simulations of impact of the superlattice on optical absorption and electron emission within the nanopillar.

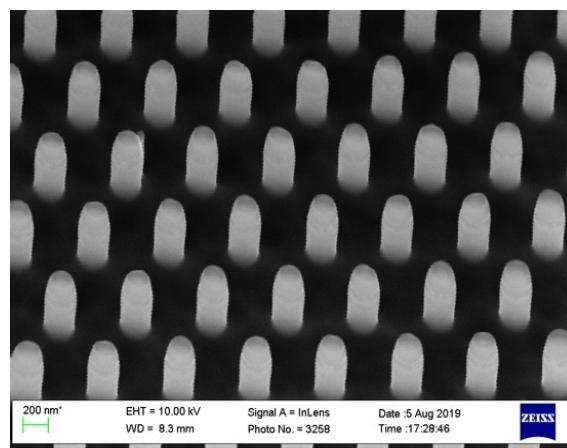


Figure 3: SEM of NPA, sidewall straightness optimized.

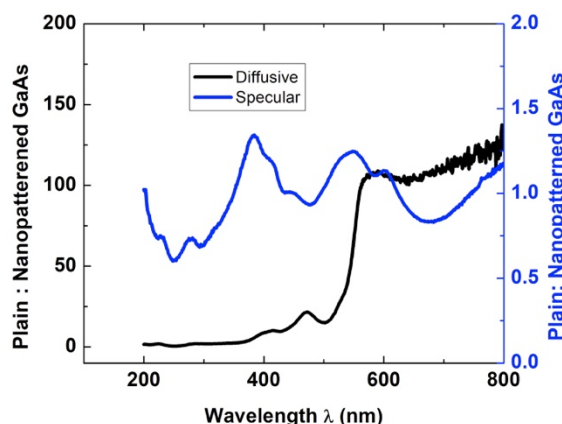


Figure 4: Spectral reflectance, NPA as a ratio with flat GaAs, specular at 8° and diffuse.

Of note, this is the first superlattice cathode grown in the US for spin-polarized photoemission in several years and represents initial steps in re-establishment of superlattice photocathode production for spin-polarized accelerators. We intend to report on simulations, integration with NPAs, and tests of resulting QE and spin-polarization via Mott polarimetry at Jefferson Laboratory in a future article. Future work includes gun testing of lifetime to assess NPA robustness under ion back-bombardment and the impact of the NPA on emittance and dark current.

CONCLUSION

We have applied Kerker conditions to Mie scattering in GaAs to provide a new geometric constraint on nanopillar size parameter. We have also achieved significant improvement in nanopillar sidewall straightness with an optimized inductively coupled plasma etch, leading to higher absorption and an expectation of higher QE alongside any lifetime improvement from back-bombardment robustness.

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