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High Polarization InAlGaAs/AlGaAs photocathodes grown using MBE

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Strained superlattices of GaAs/GaAsP grown using molecular beam epitaxy (MBE) have been used for more than 20 years to generate high polarization electron beams for nuclear physics. GaAs/GaAsP superlattices have several manufacturing challenges, including the thick graded layer required for the virtual GaAsP substrate, the significant difference in optimal growth temperatures for GaAs and GaAsP, and the scarcity of MBE systems using phosphorus. InAlGaAs/AlGaAs strained superlattice photocathode are an alternative structure that eliminates many of the hurdles to growing GaAs/GaAsP in MBE systems. Measurements of quantum efficiency and polarization from InAlGaAs/AlGaAs will be presented. These include studies on a variety of growth parameters to optimize performance, including digital alloys, growth temperature variations, and variation in structure.

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1. Introduction

High polarization electron beams are used at accelerators to explore the spin structure of nucleons. For years, the photocathode of choice for high polarization electron beams has been a strained superlattice structure of GaAs/GaAsP, where strain from lattice mismatch and quantum confinement combine to allow the preferential excitation of one spin state [1].

While GaAs/GaAsP photocathodes have proven to be a consistent material for high polarization photocathodes [2], and has been successfully grown with distributed Bragg reflectors (DBR) for high quantum efficiency [3] (QE), fabricating GaAs/GaAsP in a molecular beam epitaxy (MBE) system is challenging. Most commercial semiconductor fabrication shops do not routinely use phosphorus in their chambers; it is not used commonly in semiconductor chip manufacture or solar cells, and phosphorus residue in a system can adversely affect the growth of subsequent materials [4]. The vendor, which developed these high polarization photocathodes through a series of Small Business Innovation Research (SBIR) research projects, stopped manufacturing them ten years ago, and there is not currently a company growing GaAs/GaAsP photocathodes which will be needed for JLab, the Electron-Ion Collider (EIC) and other accelerators such as MESA at Mainz.

Jefferson Lab (JLab) and SVT Associates have collaborated on alternatives to GaAs/GaAsP, such as structures containing Sb, but polarization was not as high. Other groups have investigated InAlGaAs/AlGaAs as an alternative [5], which demonstrated polarization as high as GaAs/GaAsP, but results were inconsistent and the optimal wavelength was significantly higher than the 780 nm drive lasers for GaAs/GaAsP.

The aim of this project is to revisit the InAlGaAs/AlGaAs superlattice structure, optimizing strain, stoichiometry and growth temperatures to develop InAlGaAs/AlGaAs photocathodes with QE and polarization meeting or exceeding that of GaAs/GaAsP. Once the InAlGaAs/AlGaAs structure is optimized, our goal is to find commercial vendors to grow these structures using In and Al sources commonly found in III/V semiconductor MBE systems and re-establish a reliable source of high polarization photocathodes.

2. Strained superlattice photocathode growth

2.1 GaAs/GaAsP

Polarized electrons are emitted from a superlattice of GaAs and GaAsP layers, 3 to 5 nm thick, with between 15 and 20 pairs layered to make a photoemissive layer about 100 nm thick. The GaAs is the photoemissive layer, and when grown on a GaAsP substrate, the GaAs experiences a compressive strain that splits a degeneracy in the conduction band and allows preferential excitation of one spin state over the other [6].

A GaAs/GaAsP superlattice is grown beginning with an epi-ready GaAs substrate, then a graded transition layer is grown with phosphorous concentration varying from 0% to the desired P stoichiometry of about 35%. To grow this graded layer without dislocations or defects is difficult and time consuming, and therefore adds expense to the photocathode.

Another technical issue with GaAs/GaAsP comes from the coupling of the bandgap and the lattice constants for GaAs/GaAsP. As a binary/tertiary superlattice, once the optimal strain (phosphorous concentration) is set, the wavelength for excitation is also determined and cannot be independently adjusted.

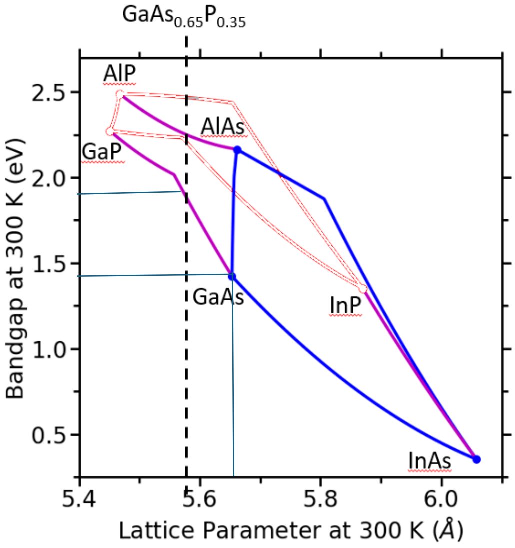

Figure 1: Bandgap vs. Lattice Parameter for GaAs/GaAsP strained superlattice photocathodes. Since all GaAsxP(1-x) barrier layers must lie on the magenta line, adjusting strain necessarily adjusts the bandgap for the material.

2.2 InAlGaAs/AlGaAs

InAlGaAs/AlGaAs has several benefits over GaAs/GaAsP from a growth standpoint. First, the lattice constant of AlAs (as well as any Al$_x$Ga$_{(1-x)}$As) is closely matched to that of the bulk GaAs substrate. This eliminates the need for a thick graded layer, reducing the time for growth and the imperfections inherent in growing the thick graded layer. In fact, a superlattice of GaAs and AlGaAs ~200 nm can replace the 2.5 um graded layer and 2.5 um GaAsP virtual substrate, cutting the fabrication time by a factor of 25.

A second fabrication benefit of InAlGaAs/AlGaAs over GaAs/GaAsP is the ability to vary the bandgap and the strain independently. Since this is a quarternary/tertiary superlattice, rather than having the strain determine the band gap, the band gap can be adjusted to match available drive laser wavelengths independently of the strain optimization. Adjusting indium concentration in the photoemissive layer changes the lattice constant and strain, allowing polarization optimization, while adjusting aluminum concentration in either the barrier or well will change the excitation wavelength for the polarization maximum.

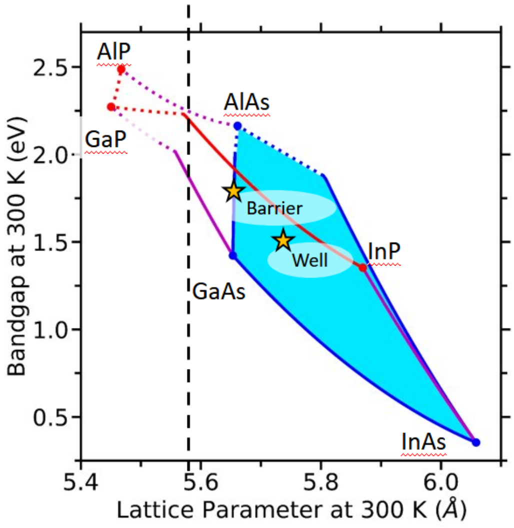

Figure 2: The quartenary alloy InAlGaAs can be anywhere in the shaded area of the graph, such that varying indium concentration will increase or decrease strain, while varying arsenic concentraion will vary the bandgap for the material.

Additionally, the optimal growth temperatures for InAlGaAs and AlGaAs are closer together than those for GaAsP and GaAs, making temperature optimization more straightforward.

Finally, distributed Bragg reflectors (DBRs) can be used to reflect light back through the photoemissive layer. For InAlGaAs/AlGaAs, the DBR is grown using a AlAs/AlGaAs and an AlGaAs cavity. The DBR structure is a subset of the sources that are already being used to grow the photoemissive layers, and the AlGaAs cavity is commonly grown in MBE systems, and has well characterized indices of refraction for different concentrations. For the GaAs/GaAsP photocathode, the DBR must be fabricated from a GaAsP/AlGaAsP superlattice, and the GaAsP cavity is less common with less information on the index of refraction, making it more difficult to optimize than the AlGaAs cavity.

3. Parametric studies of InAlGaAs/AlGaAs

3.1 Growth Temperature

The first parametric studies for the InAlGaAs/AlGaAs photocathodes were an optimization of the growth temperature while holding composition constant. The growth temperature was varied between 505 and 520 ºC, and both polarization and QE were measured in the JLab

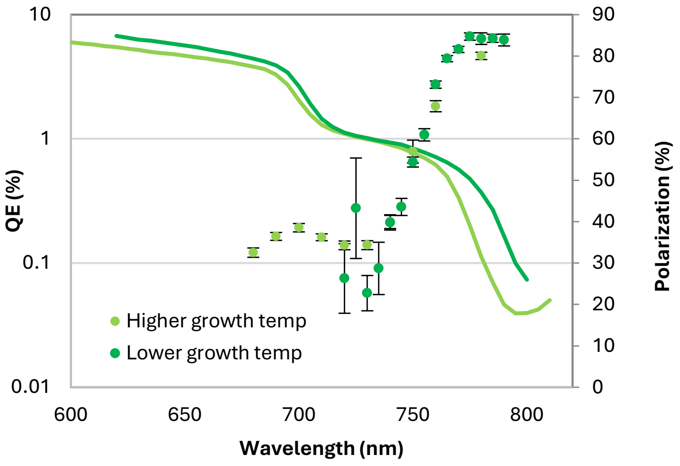

Figure 3: Growth temperature studies. The lower growth temperature appears to have higher polarization and QE, but these measurements will be repeated due to significant repairs of the microMott between the measurements.

microMott polarimeter [7]. These results suggest that the lower growth temperature yields both higher polarization and QE. However, the microMott required numerous repairs during the months between the two measurements, so this growth temperature comparison will be repeated in future.

3.2 Strain

The next study compares QE and polarization with different strain. With the quartenary/tertiary structure, strain can be tuned by varying the stoichiometry, such as increasing the indium concentration. Two different high strain photocathodes were grown, with both showing higher polarization than the initial sample tested for temperature optimization. As expected, varying the concentrations also affected the wavelength for the polarization maximum for these samples. The wavelength shift for peak polarization in the "Increase Strain 2" sample is due to a change of both the lattice mismatch and the bandgap for the material, and was fabricated in part to compare calculations of expected bandgap for a material to experimental data.

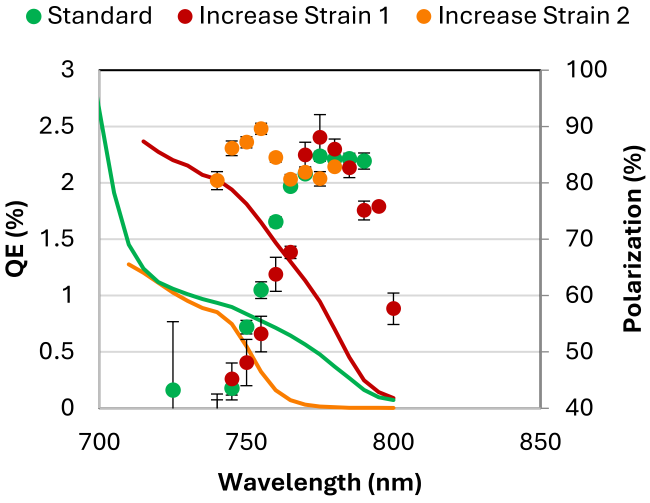

Figure 4: Strain studies find that material grown with higher strain exhibit higher polarization as expected. The wavelength shift for peak polarization was due to other parameters being tested simultaneously.

3.3 Random Alloy Disorder

The final parametric study presented here compares random alloy to digital alloy growth structures. With the quartenary photoemissive InAlGaAs layer, unlike the binary GaAs photoemissive layer in GaAs/GaAsP, the composition can be disordered, and this disorder can lead to a decrease in polarization during the electron transport to the surface [8]. To mitigate this effect in photodiodes and other structures, higher order structures like AlGaAs can be grown as alternating layers of AlAs and GaAs, with the thickness of the layers determining the stoichiometry rather than relying on the differing fluxes from each source during co-deposition [9].

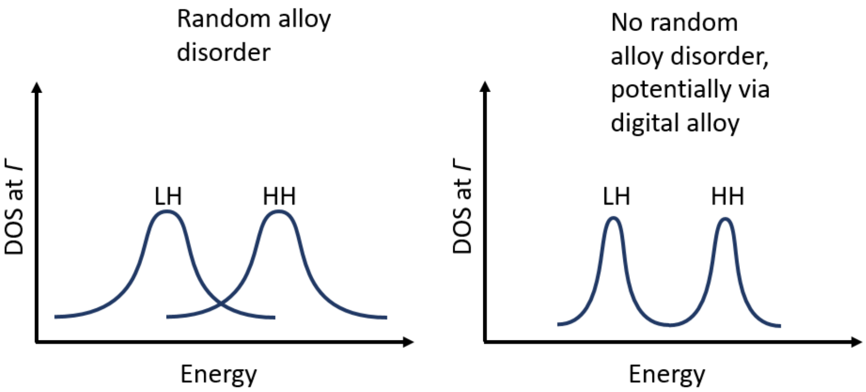

Figure 5: Random alloy disorder can broaden the light hole and heavy hole bands. Growing a digital alloy helps sharpen the heavy and light hole band densities of states, and with less overlap, there is a greater range of wavelengths for which polarization remains high.

Three digital alloy photocathodes were grown and tested. Successful narrowing of the heavy and light hole bands, yielding less overlap, should lead to a broader range of wavelengths for which polarization is at its maximum. The three digital alloy samples and the best random alloy sample are shown in Figure 6.

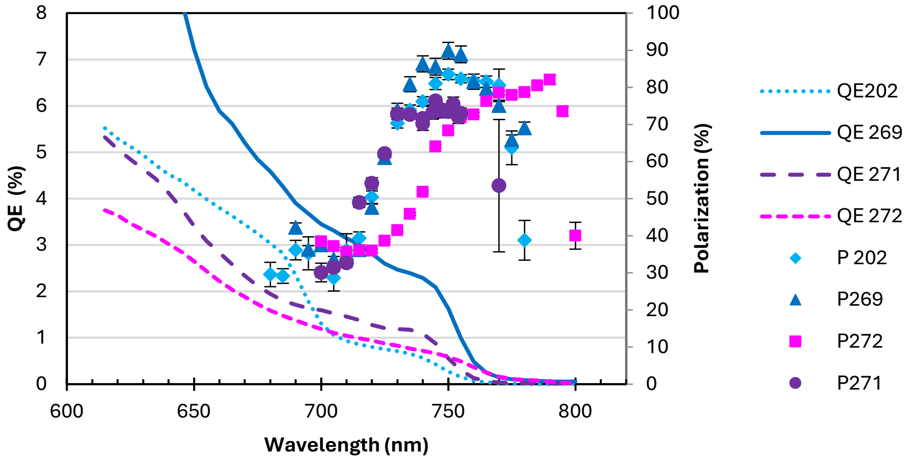

Figure 6: QE and polarization vs. wavelength for three digital alloy samples along with the best photocathode #269.

We can normalize polarization vs. wavelength data to compare the extent of the wavelength range for polarization is high for these samples. We first normalized each of the data sets to P(max), then performed a Gaussian fit for each sample's polarization data, and finally shifted the central wavelengths to overlap. Figure 7 shows the normalized fits of the data from Figure 6, with the vertical lines showing the extent of the wavelengths where polarization exceeds 90% of P(max).

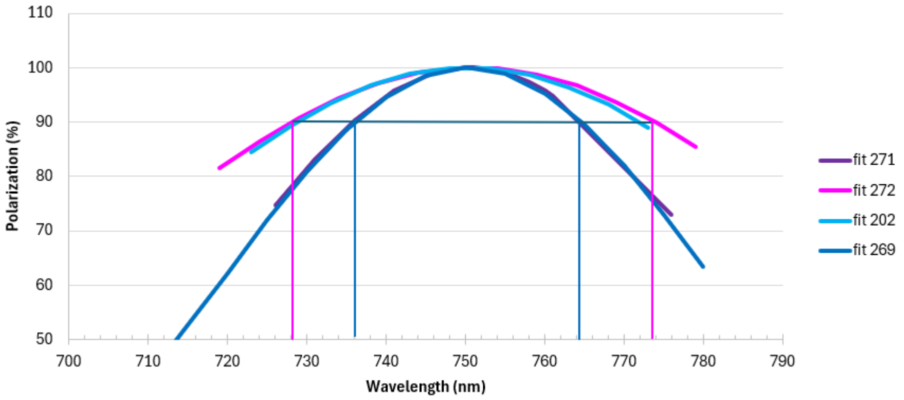

Figure 7: Normalized fits for the digital alloys and the best random alloy (Sample #269). Vertical bars show extent of wavelength for which the polarization is above 90% of Pmax. Two of the digital alloys show broader range of of high polarization, indicating better splitting of the heavy and light hole bands. The random alloy shows a narrower range for high polarization, indicating more overlap between the heavy and light hole bands.

For the random alloy, #269, the polarization is above 90% of P(max) at wavelengths from ~737 to 765 nm, giving a range of 28 nm for highest polarizations. Converting wavelength to energy, this corresponds to 62 meV splitting betwwen the heavy and light hole bands. On the other hand, for two of the digital alloys, the range of high polarization wavelengths extend from 728 through 773 nm, or about 45 nm, giving an energy of 99 meV. This indicates that the digital alloying was successful in reducing the overlap in the heavy and light hole bands for these samples. The final sample, #271, was grown to be a digital alloy but shows similar behavior to the random alloy, indicating an issue with the growth or heat cycles during measurement.

4.Conclusions

This study of InAlGaAs/AlGaAs strained superlattice photocathodes shows promise for this material as a commercially viable high polarization photocathode. It uses commonly available commercial MBE sources, has a subset of those which can be used for QE enhancement through a distributed Bragg reflector, and has well characterized optical properties for the DBR design.

These studies have been performed in a low voltage retarding field Mott polarimeter, where emission voltage is low and vacuum is worse than in our high voltage guns. We plan polarization to do polarization and lifetime studies in a high voltage electron gun in the future. Meanwhile, further parametric studies of QE and polarization will enable optimization of a InAlGaAs/AlGaAs strained superlattice photocathode, which we anticipate being able to have grown by commercial vendors, thus restoring a commercial supply high polarization photocathodes for the nuclear physics community and beyond.

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