In Pursuit of the Narrow Cone

Prospects for 2-photon and n-type Photoemission from Al_xGa_{1-x}N

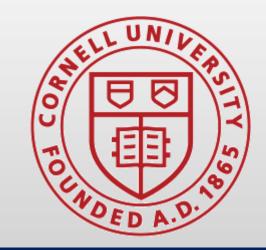
Christopher M. Pierce

Acknowledgements

Before beginning I'd like to thank all of the following people and organizations for their help on the project:

- Jared Maxson, Ivan Bazarov, and Luca Cultrera for their guidance on the project
- Howard Padmore for the inspiration to look into pump-probe photoemission
- Alice Galdi, Jai Kwan Bae, Frank Ikponmwen, Cameron Duncan, J. Kevin Nangoi for their technical assistance
- Debdeep Jena, Huili (Grace) Xing, Vladimir Protasenko, and Jimy Encomendero Risco for growing the samples used in the experiment and advice on sample preparation
- The Center for Bright Beams (NSF award PHY 1549132) for their generous funding





Outline

MTE of GaAs Photocathodes Likely Causes of MTE Growth Pump-Probe Photoemission n-Doped Photoemission **Experimental Results Future Work**

MTE of GaAs Photocathodes

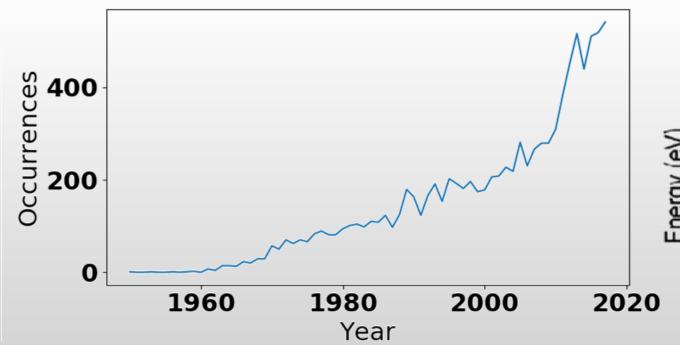
Low effective mass photocathodes like GaAs have been studied for decades

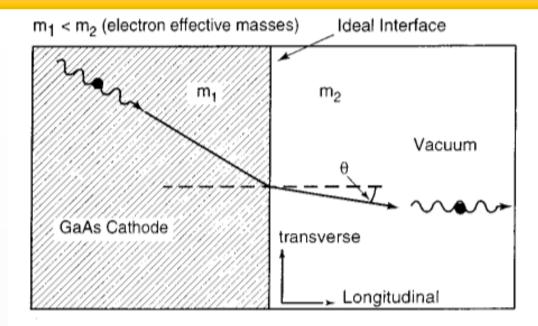
Conservation of momentum says electrons should diffract as they enter vacuum and give subthermal MTE

MTE of GaAs should be 1-2meV; outperform best photocathodes of today

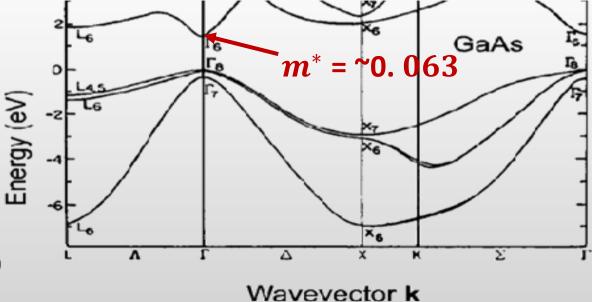
Electrons should come out in "narrow cone" of 15 degrees

Number of Articles on Google Scholar by Year for 'NEA GaAs Photocathode'





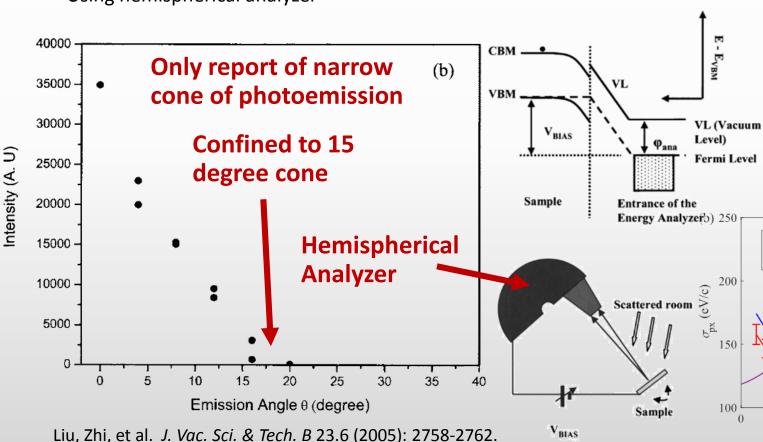
Vergara, G., A. Herrera-Gómez, and W. E. Spicer *Journal of applied physics* 80.3 (1996): 1809-1815.



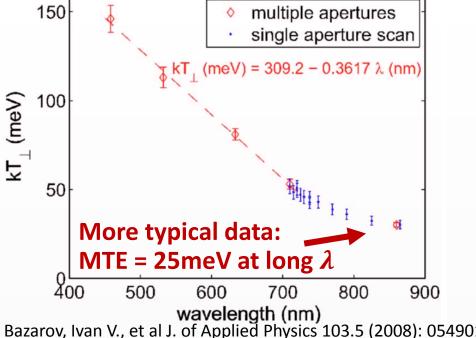
MTE of GaAs Photocathodes

Most researchers report 25meV ($k_B T_{room}$) MTE near threshold. That's over an order of magnitude greater than what's expected! Where did the MTE come from?

Exception of Z. Liu et al, who claim to see the small effective mass Using hemispherical analyzer

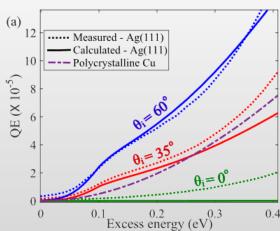


cmp285@cornell.edu



Bazarov, Ivan V., et al J. of Applied Physics 103.5 (2008): 054901.

NOTE: Transverse momentum is conserved in monocrystalline metals w/ clean surfaces.



Karkare, Siddharth, et al. Physical review letters 118.16 (2017): 164802.

0.4

Excess energy (eV)

0.3

Measured - Ag(111)

-Calculated - Ag(111)

-Polycrystalline Cu

Likely Causes of MTE Growth

Short Exposure

Long Exposure

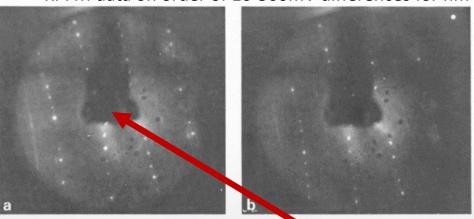
Cs NEA activation causes disorder on GaAs surface

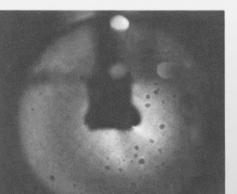
LEED patterns disappear as Cs is added

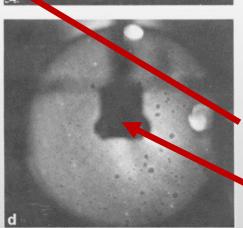
STM and DFT shows mobile layer of atoms; changes on nS times scales

Work function can be locally different than average for surface

KPFM data on order of 10-500mV differences for nm patches





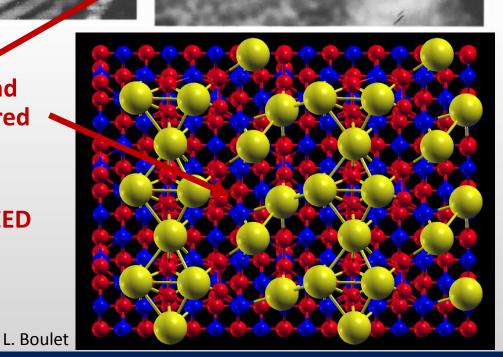


Cs Atoms move around on surface in disordered layer

Clean GaAs w/ sharp LEED

Cs NEA activation eliminates pattern

Goldstein, Bernard. Surface Science 47.1 (1975): 143-161.

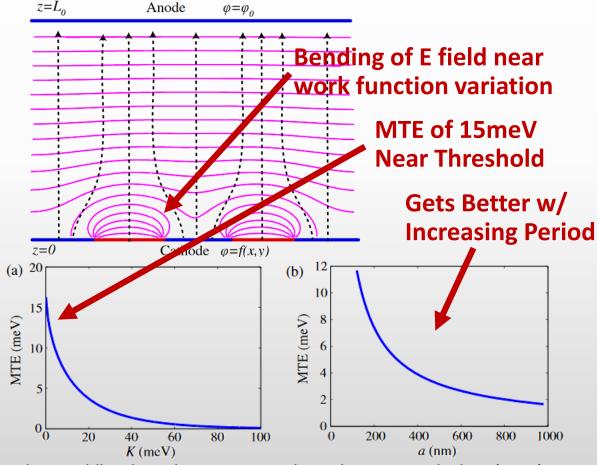


Likely Causes of MTE Growth

MTE growth can come from work function variation over small scales

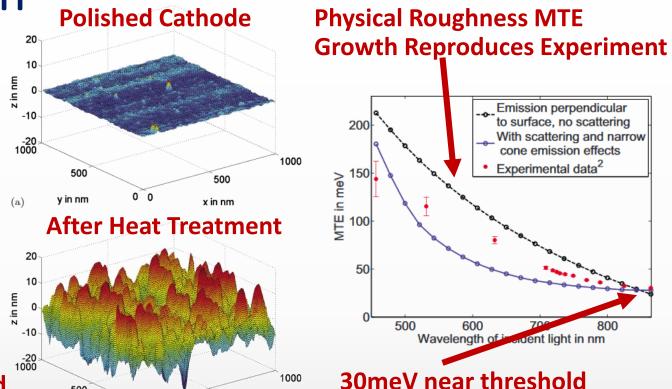
Simulations w/ realistic electron distribution and variation for GaAs show 15meV growth

Decreases w/ KE; classically, electrons spend less time in bad fields



Karkare, Siddharth, and Ivan Bazarov Physical Review Applied 4.2 (2015): 024015.

Karkare, Siddharth, and Ivan Bazarov. Applied Physics Letters 98.9 (2011): 094104.



Photocathodes also have nanoscale surface variation

AFM data shows ~10nm roughness after heat treatment on polished

Physical roughness can increase MTE

GaAs

Around 30meV based on simulation w/ data from AFM on heat treated GaAs

Scales correctly w/ wavelength, matches nicely w/ experiments

Not the whole story -> Still observe poor MTE on flat cathodes

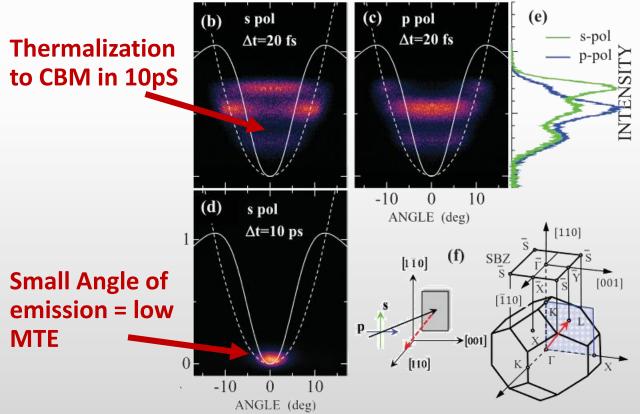
Pump-Probe Photoemission

Despite MTE growth effects, people observe momentum conservation all the time

Exploited for ARPES -> High enough excess energy to avoid surface effects

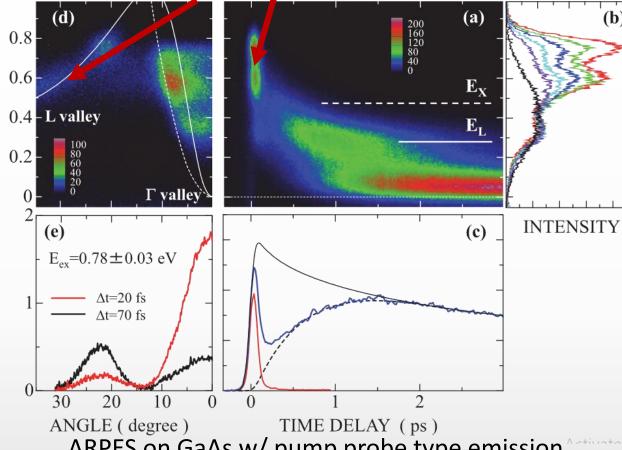
Recent paper observed narrow cone in GaAs w/ time resolved ARPES

Can we do the same to generate bright beams?



Kanasaki, Jun'ichi, Hiroshi Tanimura, and Katsumi Tanimura. Physical review letters 113.23 (2014): 237401.

Scattering to X/L Valley at large photon energy



ARPES on GaAs w/ pump probe type emission

Excite electrons into CB w/ IR->Visible light; eject from sample w/ UV

Analysis done w/ hemispherical analyzer to get energy and angle data

Observed electrons thermalize to conduction band minimum

Inter-valley scattering w/ large pump energies

(eV)

ENERGY

Pump-Probe Photoemission

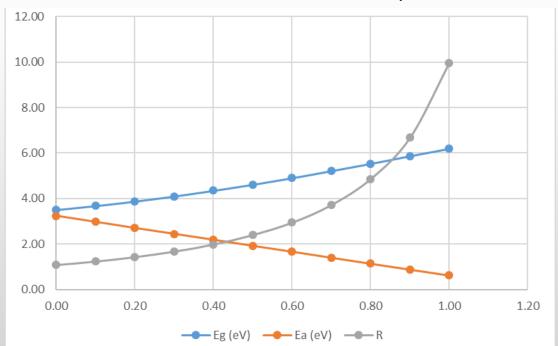
GaAs band structure leads us to believe this won't work

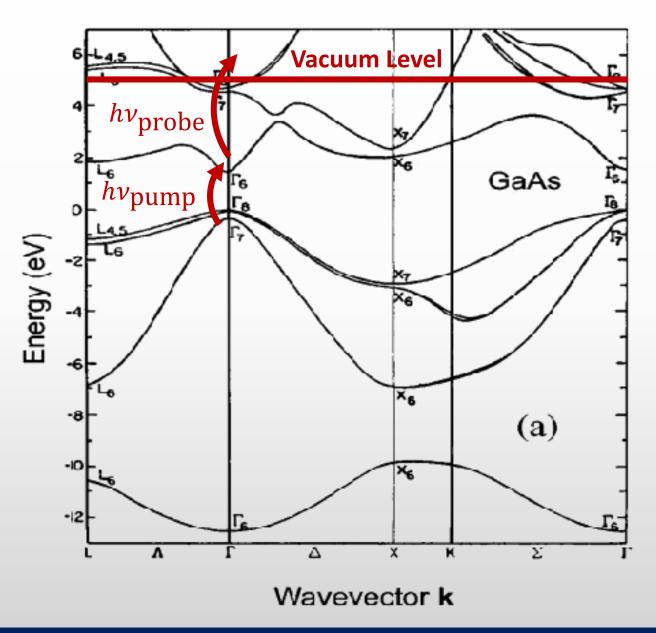
High enough energy probe photon for two photon emission

Electron-electron scattering w/ valence band In order to avoid these effects, require material w/ $E_{affinity} < E_{gap}$

Al_xGa_{1-x}N satisfies this

Tune x to a convenient value for experimental setup





n-Doped Photoemission

For initial studies, n-doped Al_xGa_{1-x}N was selected

Avoids the complication of two photon emission; carriers already in conduction band

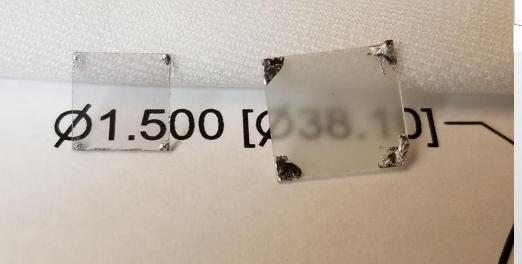
Carrier concentrations expected to be less than w/ 2 photon Has band bending, unknown how this affects QE/MTE

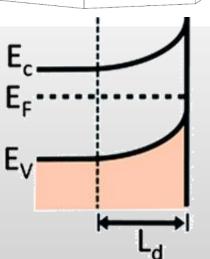
Received epitaxially grow samples from Jena/Xing group at Cornell

Grown on top of GaN on Sapphire w/ good surface quality characterized by AFM

X=0.75, Carrier Conc. = -1.058×10^{18} cm⁻³

Samples are transparent!





rms: 2.65 nm

10

Energy (eV)

100nm n-type 75%-AlGaN 16 nm AlN -→ 75%AlGaN

AM LK H

10

Al_xGa_{1-x}N (x=0.8) Band Structure

 m^*

AlN on Sapphire

Experimental Results

Sample was introduced to Cornell photocathode characterization system

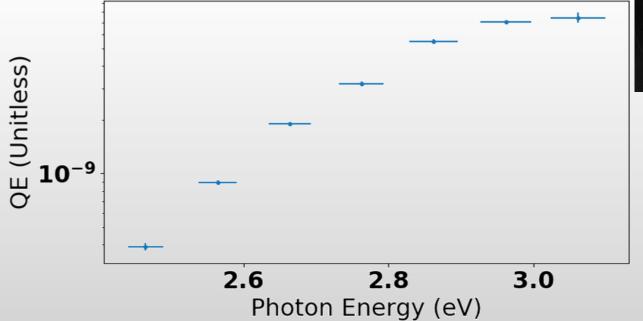
LEED showed sharp hexagonal pattern Annealed sample at 250C for 5 hours

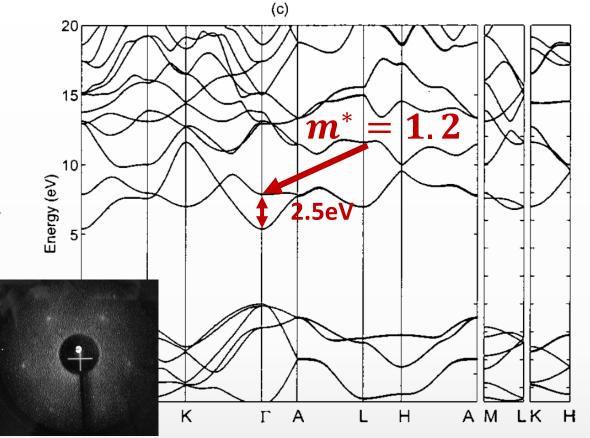
Photocurrent measurement performed on clean sample

Tunable light from supercontinuum source w/ monochromator (10nm FWHM bandwidth; 10s of pS pulse length); ~1mW/nm power

Lock-in amplifier + chopper used to measure photocurrent of cathode biased to -18V. (saw less than nA current)

Not enough current for MTE measurement





This led us to activate the sample by cesiation

Hope to get high enough QE to perform MTE measurement

Could cause issues w/ MTE growth effects as before

Still good reason to believe good MTE is achievable Many surface effects are diminished at high excess energy Emission from conduction band allows us to use large photon energy.

11

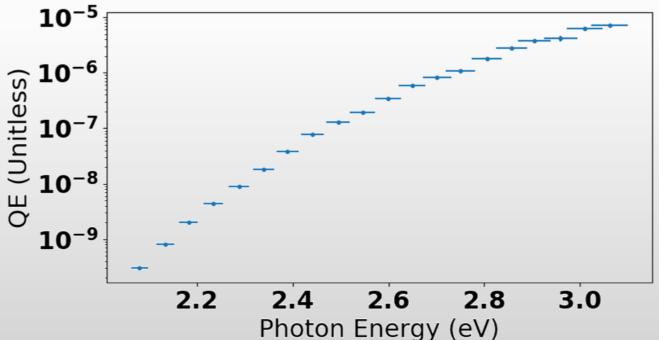
Still have to watch out for band w/ large m^*

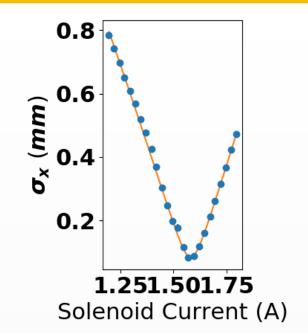
Experimental Results

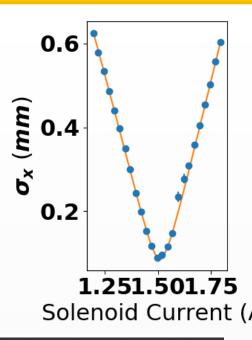
Recorded two order of magnitude increase in QE Still have threshold of ~2eV, so we aren't NEA

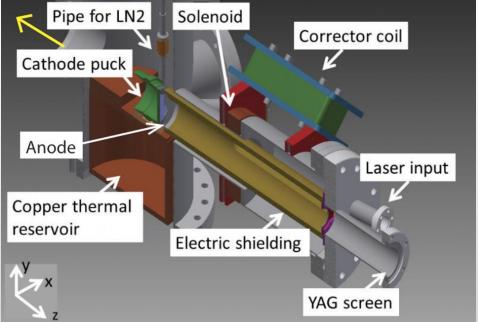
Performed MTE measurements at variety of photon energies

Same supercontinuum source as before Cornell TE meter at 10Kv, performing solenoid scans Sensitive to beam currents < 1nA near beam waist









Lee, Hyeri, et al. Review of Scientific Instruments 86.7 (2015): 073309.

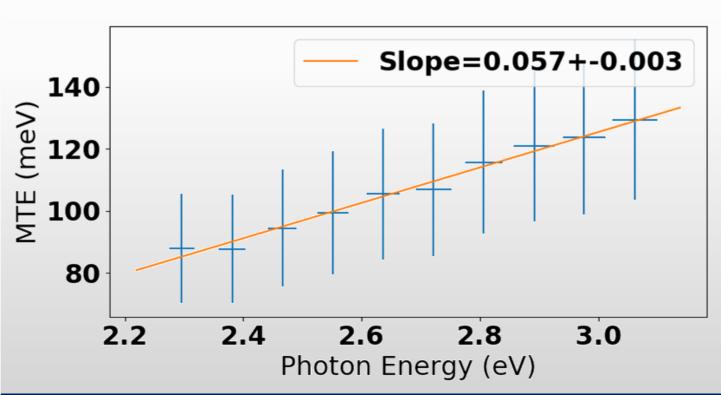
Experimental Results/Future Work

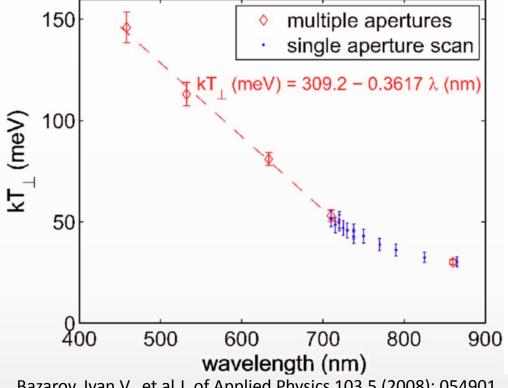
MTE has some interesting features to it

Still far above 5meV expected from $m^* = 0.2$

No jump in MTE at 2.5eV where $m^* = 1.2$ band is located

Increases w/ slope of ~0.06, much less than value for disordered photocathodes





Bazarov, Ivan V., et al J. of Applied Physics 103.5 (2008): 054901.

Search is not over, still must investigate pump-probe photoemission

Simulations show an order of magnitude better carrier concentration

Avoids issues with cesiation of surface No band bending as in n-doped samples