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# MEASURING THE MEAN TRANSVERSE ENERGY OF PUMP-PROBE PHOTOEMITTED ELECTRONS

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*Abstract*

Low effective mass semiconductor photocathodes have historically failed to exhibit the sub-thermal mean transverse energies (MTEs) expected of them based on their band structure. However, conservation of transverse momentum across the vacuum interface, and therefore a low MTE in these materials, has been observed in time resolved ARPES. To help bridge this gap, we measured the MTE of the pump probe photoemitted electrons seen in the ARPES experiment using methods typical of accelerator physics. We compare the results of these measurements with those of both communities and discuss them in the context of photoemission physics.

## INTRODUCTION

The discovery of new low MTE photocathodes is a requirement for those seeking to improve the brightness of photoinjectors operating at the charge extraction limit. In photoinjectors with a fixed electric field there is a minimum initial spot size $\sigma_{x,\min}$ for every bunch charge due to the virtual cathode instability. This is called the charge extraction limit. Since the charge extraction limit sets a maximum charge density in the spatial coordinate and cathode MTE sets the density in the momentum coordinate, the maximum brightness can be written as $B_{4D,\max} \propto (\sigma^2_{x,\min}\text{MTE})^{-1}$. The only way to improve the brightness of photoinjectors operating at or near $\sigma_{x,\min}$ is to drive down the MTE of photocathodes.

### *Low Effective Mass Semiconductor Photocathodes*

Low effective mass semiconductors offer a promising path towards new low MTE photocathodes. Conservation of transverse momentum across the photocathode's surface means that the transverse energy of electrons is scaled by the mass ratio $m_e^*/m_e$ upon emission. For near threshold photoemission from a negative electron affinity (NEA) photocathode, the transverse energy of electrons comes from the Fermi tail and is $k_BT$. The MTE is then $(m_e^*/m_e)k_BT$ and makes us interested in candidate photocathodes with small $m_e^*$. Some semiconductors, such as GaAs, have an effective mass ratio as small as $m_e^*/m_e = 0.07$ and should have MTEs as low as 1.7 meV for near threshold photoemission at room temperature [1]. Compare this with the MTE of polycrystalline copper, a commonly used metallic photocathode, which has been measured at 85 meV near threshold [2].

The MTE of NEA GaAs near threshold has been consistently measured at more than 25 meV which is $k_BT$ at room temperature [3–8]. There is only one reported measurement of the expected 1.7 meV MTE and it has not been reproduced since [9]. There is currently no consensus on why the MTE is so much larger than what the material's low effective mass would lead us to believe. Some proposed explanations include physical/chemical roughness of the surface, scattering of electrons in the Cs overlayer, and the effects of electron-phonon scattering [1, 10, 11].

The narrow dispersion of the GaAs conduction band is observed in time resolved ARPES (trARPES). In a trARPES measurement, electrons are excited into empty conduction band states by pump photons and emitted into the vacuum some time later with probe photons. These photoelectrons are filtered by angle of emission and longitudinal energy in a hemispherical analyzer. Kanasaki et al. measured a photoelectron distribution from GaAs which can be naively converted to a transverse energy distribution with 1.7 meV MTE [12]. Photoinjectors, however, do not have the energy filtering capabilities of an ARPES style detector. An accurate measurement of MTE for these applications must include the full photoelectron distribution, not a narrow subset of it.

## MEASUREMENTS OF THE OUT OF EQUILIBRIUM PHOTOCATHODE

The photocathode was prepared by solvent cleaning and etching a p-type GaAs (110) wafer obtained from a commercial source. The wafer was Zn doped to a p-type carrier concentration of $1 \times 10^{19}$ cm$^{-3}$ and came polished to a surface roughness of better than 0.4 nm RMS. Before introduction to a vacuum chamber at better than $1 \times 10^{-10}$ Torr pressure, the wafer was cleaned in acetone and etched in a 1% HF solution for 30 s. Inside the preparation chamber, the sample was annealed at 550 °C for 8 hours to remove surface oxides. The band gap was measured at 1.40 eV using photoluminescence spectroscopy with illumination at 633 nm. We estimate the workfunction of the sample to be 4.6 eV based off of measurements of quantum efficiency (QE) as photon energy was changed. The data, in Fig. 1, was fit to a model with quadratic dependence of QE on excess photon energy.

An optical parametric amplifier was used to generate the 750nm pump and some of the 1030 nm output of an Yb fiber laser driving it was diverted and frequency tripled for use as the probe. Both were overlapped at the sample and aligned in time of arrival. The intensity of the pulses was increased to the point that each caused multiphoton photoemission from the sample and those multiphoton beams were used to center the laser spots on the cathode and align them with each other. The cross correlation of the pulses was measured as less than 400 fs full width at half maximum by watching

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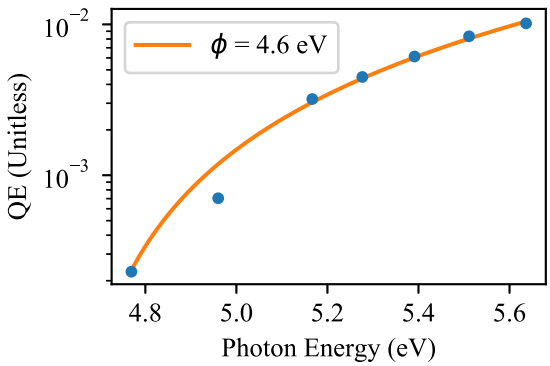


Figure 1: Quantum efficiency (QE) as a function of photon energy for the GaAs (110) sample. Photocurrent was measured at five intensities for each photon energy to accurately determine the QE and to ensure there was no multiphoton photoemission. A quadratic model, shown in orange, was fit to the data with workfunction ($\phi$) as a fit parameter. The fit value of the workfunction was $\phi = 4.6$ eV.

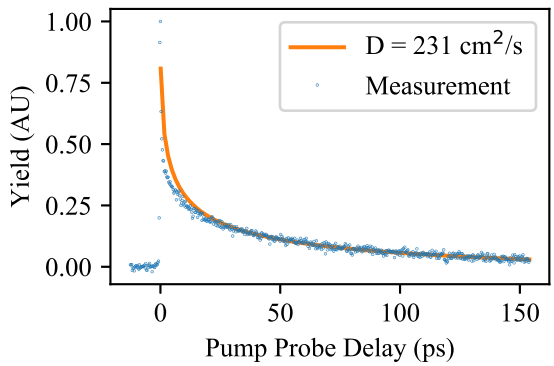


Figure 2: In blue, measurements of photocurrent are displayed as pump probe delay ($\Delta t$) is changed. The large peak at $\Delta t = 0$ is due to the coherent enhancement of multiphoton photoemission from the pump and probe. The long tail afterwards is 1+1 photoemission of electrons being excited into the GaAs conduction band. The curve in orange is a model including carrier recombination and diffusion with a fit value of the diffusion constant. We found an optimal value of the diffusion constant to be 231 cm$^2$/s. Values of the absorption length for the pump and probe are from the Handbook of Optical Constants of Solids [13]. A constant multiphoton background was subtracted from the signal and the result normalized to a peak value of one before fitting.

the coherent enhancement of multiphoton photoemission as the time delay of the pulses was varied.

### *Yield and Carrier Diffusion*

We measured photocurrent as a function of pump probe delay and confirmed the presence of long lived carriers in the conduction band of the sample. Our measurement is shown in Fig. 2. The time axis is signed so that there is positive delay when the probe comes after the pump. Coherent multiphoton photoemission is seen in the few hundred femtoseconds around $\Delta t = 0$. Incoherent multiphoton photoemission follows that at positive values of $\Delta t$ and extends beyond 100 ps. The number of electrons emitted in this process is proportional to how many excited carriers remain in the region of photoemission after the pump probe delay.

The time dependence of the yield curve is not explained by carrier recombination alone and an accurate description needs to include the effects of carrier diffusion. The minority carrier lifetime of p-type GaAs is 4 ns [14] and one would naively expect the lifetime of the 1+1 photoemission signal to be similar. Modeling the system to include the 1D diffusion of electrons away from the surface gives a more accurate time dependence when using literature values of the diffusion constant and of the absorption lengths for the pump and probe photons. The yield measurement as well as a fit with the diffusion constant as the free parameter are shown in Fig. 2. The fit value of the diffusion constant was 231 cm$^2$/s compared to other works which found it to be 200 cm$^2$/s [15].

### *Mean Transverse Energy*

The MTE of the out of equilibrium photocathode was measured at 200 meV for large pump probe delays. Voltage scan measurements were performed using voltages between 2 kV and 10 kV as the pump probe delay was increased in 200 fs steps away from the overlap position up to a maximum delay of 20 ps. A custom microchannel plate and scintillator based detector was coupled with an intensified CCD camera to measure the size of the low current beam. Before MTE measurements were performed, the intensity of the pump and probe were decreased to the point that no multiphoton photoemission from the individual pulses was seen in beam images at negative pump probe delays. A 100 µm diameter pinhole in the path of the probe was imaged onto the cathode with unity magnification. The MTE, shown in Fig. 3 as a function of pump probe delay, relaxes on a picosecond timescale from nearly 400 meV in the overlap region to 200 meV at large delays. There was an asymmetry in the MTE measurement between the X and Y projected values on the scale of 4% which is attributed to an artifact of performing a measurement of such a large MTE in an instrument designed for studying low MTE photocathodes.

We repeated the time resolved MTE measurement as the intensity of the pump and probe were varied with no change in MTE at large delays. Multiphoton excitation within the individual pump and probe pulses could inflate the observed MTE by exciting electrons into unwanted states with large transverse energy. The rate of this process depends on intensity in a non-linear way which makes the MTE change with intensity if multiphoton excitation is present. We measured no change in the MTE as pump intensity was varied and only a small change in MTE, as shown in Fig. 4, during the coherent overlap when the probe intensity was changed. The bunch charge of the emitted pulse also depends on pump and probe intensity and the fact that the measured value did not

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change with bunch charge indicates that the measurement was not affected by space charge.

The measurements were performed as the pump wavelength was changed between 700 nm and 800 nm with no change in MTE. The GaAs L valley is only 300 meV above the Γ valley and so excitation with wavelengths of light shorter than 725 nm can transition electrons into states with large transverse momentum. As our pump wavelength crossed the 725 nm threshold there was no change in MTE at any pump probe delay. This is evidence that L valley electrons are not contributing to the large MTE measured.

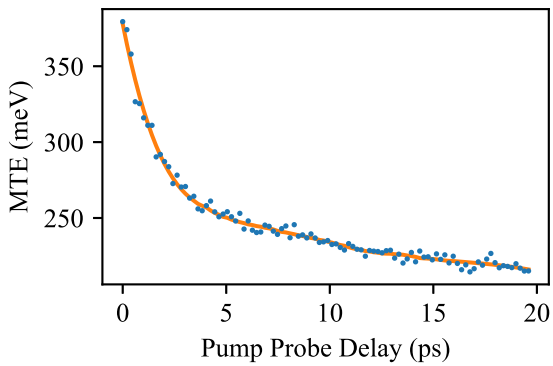


Figure 3: MTE of the pump probe photoemitted electrons as pump probe delay is changed. A smoothed curve, orange, is included to help guide the eye.

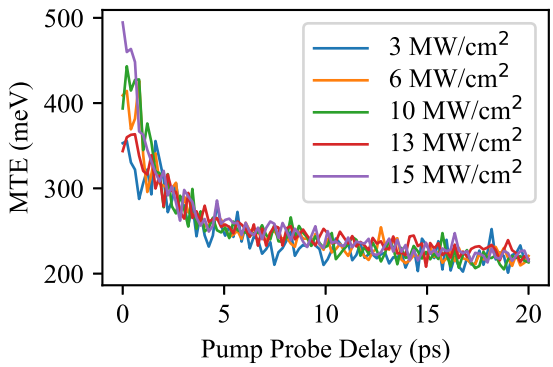


Figure 4: MTE as pump probe delay is changed at different probe intensity. Pump intensity was held at a constant 2 MW/cm$^2$. The MTE does change by a small amount, but only for delays where the pump and probe overlap each other.

Adsorbates are not a likely cause of the observed large MTEs because the measurements were replicated immediately following annealing and were uniform across the cathode. The cathode was annealed at 550 °C for 8 hours to remove adsorbates and immediately transferred to the MTE meter where measurements were performed within 15 minutes of shutting off the heater. The vacuum in the chamber was such that the time to form a monolayer of contaminants was several hours. Measurements were repeated in half hour increments following transfer to the MTE meter with no change observed. The MTE was determined to be uniform across the cathode by repeating the time resolved measurement at ten different locations across the surface.

## DISCUSSION

We have measured the MTE of pump probe photoemission from the GaAs (110) surface to be two orders of magnitude larger than what the low effective mass argument would lead us to believe. Although the low effective mass dispersion was seen in trARPES, this does not contradict our new measurement. The trARPES experiment does not collect the full distribution of electrons emitted from the sample and there could be electrons outside of the narrow energy acceptance of the detector that contribute to a large MTE. We did observe relaxation of the transverse momentum distribution on a timescale that is longer than the overlap of the pump and the probe. This is likely thermalization of the electrons as they make their way to the bottom of their band by non-radiative transitions.

Estimates show that the dispersion of the GaAs (110) surface state is wide enough to be a potential cause of the larger than expected MTE we have observed. Ivanov et al. calculated the surface band structure of GaAs (110) and found it to have states immediately below the conduction band minimum of the bulk [16]. The probe used in our experiment has enough energy to emit electrons in these states and the wide dispersion of their band means that the photoelectrons can have large transverse momentum. These electrons would also have a small enough longitudinal momentum that they would not show up in the trARPES experiment which only focused on emission from the conduction band. Confirmation of this theory would require an angle and energy resolved measurement of the full electron distribution.

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