

ULTRAFAST NONLINEAR PHOTOEMISSION FROM ALKALI ANTIMONIDE PHOTOCATHODES*

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Abstract

Alkali antimonides photocathodes are a popular choice of electron source for high average brightness beams, due to their high quantum efficiency (QE) and low mean transverse energy (MTE). This paper describes the first measurements of their nonlinear photoemission properties under sub-ps laser illumination. These measurements include wavelength-resolved power dependence, pulse length dependence, and temporal response. The transition between linear and nonlinear photoemission is observed through the wavelength-resolved scan, and implications of nonlinear photoemission are discussed.

INTRODUCTION

High brightness photoinjectors are an integral part of ultrafast electron diffraction (UED) and free electron laser (FEL) beamlines. For both UED and FELs, lower emittance and shorter bunches contribute to better performance. The lowest emittance achievable in a beamline is constrained by Liouville's theorem to be the emittance of the beam immediately on birth, while bunch length is determined primarily by the pulse length of the laser used to photoemit the electrons as well as any downstream compression optics. The maximum brightness of a photoinjector, defined as current divided by emittance, is determined solely by the mean transverse energy (MTE) of the photocathode and the accelerating gradient. Due to space charge effects near the cathode, there is a fundamental tradeoff between having low emittance beams and having ultrashort beams.

Thus, one clear path for improving the quality of electron beams in photoinjectors is decreasing the MTE of the photocathodes. Alkali antimonides are a class of photocathodes that have not only relatively low MTEs, but also have high quantum efficiency (QE), requiring fewer photons to produce the same number of electrons. For ultrafast, high power applications, this can be critical for lowering the laser power requirements to practical levels.

However, in 1964, Spicer [1] discovered nonlinear photoemission, where multiple photons transfer energy to an electron and emit it, in exactly this class of materials. Nonlinear photoemission causes the effective MTE to increase drastically, as the MTE is linear with photon energy [2], and the photon energy is effectively multiplied by the number of photons that participated in the photoemission. At short wavelengths, the single photon energy is large enough that

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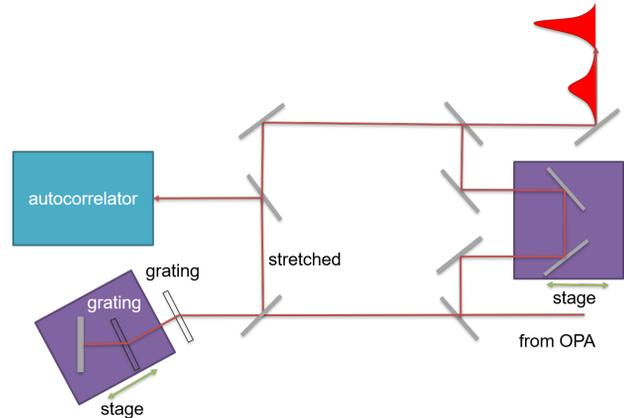


Figure 1: Schematic of the optics setup used for this experiment. The laser from the OPA is split into two different branches. One is sent through a delay stage, while the other is sent into a pulse stretcher/compressor to vary the pulse length. This is measured with a downstream autocorrelator before the pulses are recombined and sent into the gun.

linear photoemission is the dominant phenomenon. However, in order to reduce MTE, generally, it is preferred to use photon energies near threshold, which has the consequence of reducing QE. As the linear photoemission becomes weaker, nonlinear photoemission is responsible for a larger proportion of the emitted electrons. This can cause the MTE to actually begin to increase as the photon energy is reduced, as has been calculated for copper [3].

In this paper, we describe the first measurements of ultrafast nonlinear photoemission phenomena from alkali antimonides. These were focused on measuring the photoemission yield to determine the photon energies at which nonlinear photoemission begins to become measurable and the proportion of electrons that are emitted in this fashion.

EXPERIMENTAL SETUP

The experimental apparatus consisted of two main components, the laser and the beamline. The laser was a commercially available Yb fiber laser coupled to an optical parametric amplifier (OPA), manufactured by Amplitude/APE. An OPA was used to generate wavelength-tunable sub-ps pulses. Typical pulse lengths are around 300 fs, and the laser can be tuned from UV to IR wavelengths with very little difficulty.

The light from the OPA is split into two legs, as shown in Fig. 1. One leg is sent through a delay stage, while the other is stretched by a double-grating pulse stretcher before

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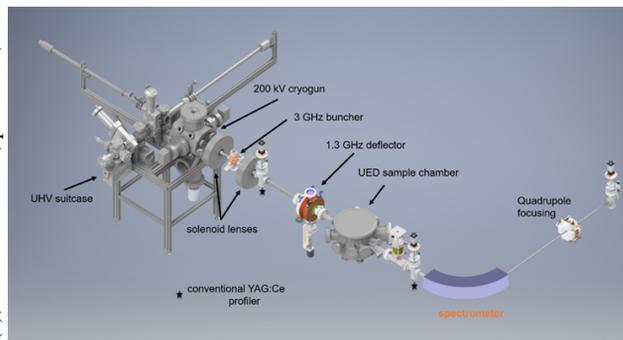


Figure 2: A model of the beamline which will be used for future experiments. The primary components consist of a 200 kV dc cryogun, two solenoid lens, bunching and deflecting rf cavities, several diagnostic YAG screens, a diagnostic/sample chamber, and a spectrometer. As of now, the components on the beamline consist of the gun, solenoids, YAG screens, and chamber, which allow us to fully map the 4D transverse phase space. The longitudinal phase space control and diagnostics will be added at a later date.

the two are recombined and sent into the electron gun. This allows us to vary both the overlap of the two pulses and the length of the pulse from the fixed leg. The amount of stretching can be measured by an optical autocorrelator in the fixed leg. The ability to change the temporal properties of the two pulses enables the measurement of the lifetime of the excited states that the electrons are emitted from. Two-photon photoemission can occur either coherently, where both photons are simultaneously absorbed by an electron, or incoherently, where a photon excites an electron into a higher energy state, where it is subsequently emitted by a different photon. If incoherent photoemission is occurring, varying the delay between the pulses would enable us to find the lifetime of these transient excited states.

The electron gun used for the experiment was a 200 kV dc gun [4] with an NaKSb photocathode. For these particular measurements, the gun was run at 50 kV and room temperature, and a Faraday cup at the end of the beamline was used to measure current, as photoemission yield was the primary focus.

Figure 2 shows a diagram of the planned future beamline. Although the measurements described in this paper only involved the gun and the Faraday cup, the beamline currently also has the two solenoids, the screens, and the diagnostic/sample chamber. The diagnostic/sample chamber contains two slits, as in [5]. This will enable us to map out the full 4D transverse phase space, and measure emittance and MTE both directly and with the solenoid scan method described in [6].

EXPERIMENTAL RESULTS

The experiment itself was conducted in two parts, measurement of photoemission yield and measurement of the lifetime of the excited emitting states. The photoemission yield was measured by measuring the current on a Faraday

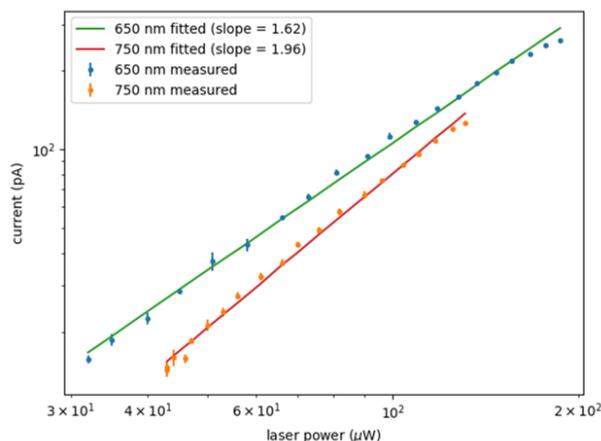


Figure 3: Onset of nonlinear photoemission from NaKSb. The power law dependence of extracted current yield on the applied laser power was measured, keeping the laser spot size and pulse length constant. At 650 nm, the slope is between 1 and 2, indicating partial two-photon photoemission. At 750 nm, the slope is almost exactly 2, indicating that linear photoemission has ceased and the photoemission is entirely two-photon.

cup in the beamline. For a given wavelength, the power was varied using a waveplate and polarizer, and the current was measured at each power. This was done for several wavelengths to measure when the transition occurred between linear and nonlinear photoemission.

We found that at 650 nm, a photon energy of 1.9 eV, close to the previously measured threshold energy of 1.8 eV [7, 8], the linear photoemission and nonlinear photoemission are both noticeably present, as seen in Fig. 3. The slope of the line on a log-log plot is approximately 1.6. Contrast this to the slope of 1.96 measured at 750 nm, corresponding to a photon energy of 1.65 eV, well below threshold. The slope here indicates nearly pure two-photon photoemission. This is consistent with the photon energy at this wavelength being too small to release electrons from the material, while doubling the energy with a two-photon process is still more than enough to release the electrons.

In addition, to confirm that the yield at 750 nm was quadratic with peak power, we measured the dependence of the yield on the laser pulse length. If the photoemission is a two-photon process, we expect that the relation $I_{avg} \propto (\frac{E}{\tau})^2 \tau$ would hold, where I is the average current, F is the laser fluence, and τ is the laser pulse length. This is because I_{avg} is linear with bunch charge, which is equal to $I_{peak} \tau$, and $I_{peak} \propto (\frac{E}{\tau})^2$ for two-photon photoemission. Thus, we expect that the yield would go as $1/\tau$.

Figure 4 shows the measured relation between yield and pulse length. A $1/\tau$ dependence was a poor fit for the data at the nominal pulse lengths, but with a 600 fs offset in the pulse length, the data is fit well by a $1/\tau$ relation. The offset is due to an expected pulse front tilt from both the double grating stretcher and the 45 degree angle of incidence on the

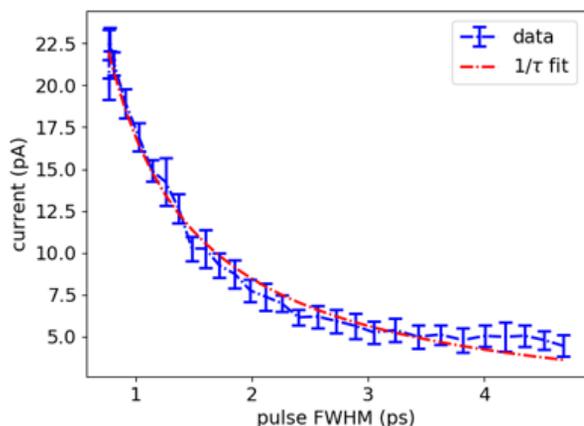


Figure 4: Measurement of the dependence of photoemission yield on pulse length at 750 nm from NaKSb. If two-photon photoemission is the dominant process, we expect a $1/\tau$ dependence, which we observe. The dependence measured here is thus fully consistent with the previous measurement of a nearly quadratic power dependence at this wavelength.

cathode. With this offset, the pulse length data is also fully consistent with a purely two-photon photoemission process at 750 nm.

The second half of the experiment was to measure the lifetime of the excited states that are being photoemitted from. We did this by comparing the autocorrelator trace from measuring the SHG signal from a BBO crystal to the trace from measuring the electron signal from a photocathode.

The optical autocorrelator consisted of a beamsplitter and a mirror mounted on an oscillating stage. By connecting a UV photodiode to an oscilloscope, we were able to measure the shape of the laser pulse directly through the increase in SHG signal when the pulses were overlapped. The result is shown in Fig. 5 in the blue line.

The charge autocorrelation measurement was done using the optical setup mentioned above and shown in Fig. 1. By varying the position of the delay stage and measuring the current at each position, we were able to map out the pulse shape just as for the optical one. The results are shown in the orange points in Fig. 5.

The results from the two autocorrelation methods are very similar. If long-lived incoherent processes were a substantial contributor to the overall nonlinear photoemission, the charge autocorrelation should be broadened. This is due to the fact that the characteristic timescale of a long-lived incoherent process is not the pulse length but the lifetime of the transient excited states. Instead, since the charge autocorrelation is not broadened, we conclude that any incoherent processes, if they exist, cannot be long-lived.

It is surprising that we were unable to find long-lived incoherent processes, as the bandgap is only 1.1 eV and the electron affinity is 0.7 eV [8], so the first pulse is sufficiently energetic to pump electrons into the conduction band, from which the second pulse could emit electrons. It appears that

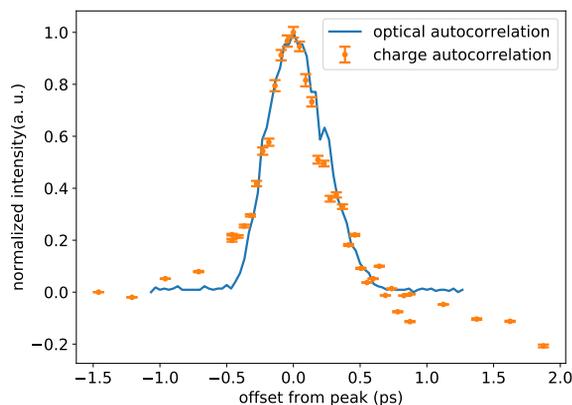


Figure 5: Promptness of nonlinear photoemission from NaKSb at 750 nm. The laser pulse shape was measured by optical autocorrelation and compared to the temporal photoemission response from the cathode. The optical autocorrelation consisted measuring the dependence of the SHG response in a BBO crystal on the delay between the two optical paths, while the charge "autocorrelation" consisted of measuring the dependence of the photoemitted current on delay. The two methods give very similar traces, indicating that there are no long-lived processes, and thus, the photoemission is prompt and coherent.

the lifetime of the conduction band states is less than the pulse length of our laser, in contrast to other semiconductors such as GaAs, where there are long-lived incoherent processes [9].

FUTURE WORK

The next planned step in this work is to measure, not only the electron yield, but also the dependence of MTE on photon energy with ultrashort pulses. In addition, we are planning to cryocool the gun and measure the dependence on MTE on the cathode temperature. In principle, MTE is limited by kT , the thermal energy of the electrons in the cathode. By reducing the temperature of the cathode, a smaller MTE should be achievable [10].

CONCLUSION

In conclusion, the first characterization of the onset of nonlinear photoemission in alkali antimonide photocathodes from an ultrafast laser has been performed. Substantial nonlinearity is already seen near threshold at 650 nm, and nearly complete nonlinearity is seen at 750 nm. In addition, no evidence of long-lived incoherent processes was seen in the autocorrelation traces. Future work will involve measuring the MTE to determine the optimal photon energy to use, as well as to determine the brightness limits that nonlinear photoemission puts on this class of photocathodes.

REFERENCES

- [1] H. Sonnenberg, H. Heffner, and W. Spicer, "Two-photon photoelectric effect in Cs_3Sb^1 ," *Appl. Phys. Lett.*, vol. 5, no. 5, pp. 95–96, Sep. 1964.
- [2] I. Bazarov *et al.*, "Thermal emittance measurements of a cesium potassium antimonide photocathode," *Appl. Phys. Lett.*, vol. 98, no. 22, p. 224101, May 2011.
- [3] J. K. Bae, I. Bazarov, P. Musumeci, S. Karkare, H. Padmore, and J. Maxson, "Brightness of femtosecond nonequilibrium photoemission in metallic photocathodes at wavelengths near the photoemission threshold," *J. Appl. Phys.*, vol. 124, no. 24, p. 244903, Dec. 2018.
- [4] H. Lee, X. Liu, L. Cultrera, B. Dunham, V. O. Kostroun, and I. V. Bazarov, "A cryogenically cooled high voltage DC photoemission electron source," *Rev. Sci. Instr.*, vol. 89, no. 8, p. 083303, Aug. 2018.
- [5] C. Gulliford *et al.*, "Demonstration of low emittance in the Cornell energy recovery linac injector prototype," *Phys. Rev. ST Accel. Beams*, vol. 16, no. 7, p. 073401, Jul. 2013.
- [6] I. V. Bazarov *et al.*, "Thermal emittance and response time measurements of negative electron affinity photocathodes," *J. Appl. Phys.*, vol. 103, no. 5, p. 054901, Mar. 2008.
- [7] L. Cultrera *et al.*, "Growth and characterization of rugged sodium potassium antimonide photocathodes for high brilliance photoinjector," *Appl. Phys. Lett.*, vol. 103, no. 10, p. 103504, Sep. 2013.
- [8] C. Ghosh and B. P. Varma, "Preparation and study of properties of a few alkali antimonide photocathodes," *J. Appl. Phys.*, vol. 49, no. 8, pp. 4549–4553, Aug. 1978.
- [9] C. M. Pierce, I. V. Bazarov, L. Cultrera, J. Maxson, "Measuring the mean transverse energy of pump-probe photoemitted electrons," presented at NAPAC'19, paper MOZBB6, this conference.
- [10] L. Cultrera, S. Karkare, H. Lee, X. Liu, I. Bazarov, and B. Dunham, "Cold electron beams from cryocooled, alkali antimonide photocathodes," *Phys. Rev. ST Accel. Beams*, vol. 18, no. 11, p. 113401, Nov. 2015.