ISBN: 978-3-95450-219-6 ISSN: 2226-0366 doi: 10.18429/JACoW-LINAC2024-MOZA002

STATE-OF-THE-ART PHOTOCATHODES FOR BRIGHT-BEAM AND SPIN-POLARIZED-BEAM GENERATION

Oksana Chubenko*, Northern Illinois University, DeKalb, IL, USA

Abstract

Due to their key properties, photoemission-based electron sources (photocathodes) are now widely used to generate bright and spin-polarized electron beams for advanced accelerator applications. The performance of these applications is limited by the quality of the photocathodes. Therefore, research and development of high-quality photocathodes with superior brightness and, where required, spin-polarization that can endure realistic photoinjector conditions is crucial. This paper will cover some of the latest advancements in photocathodes for generating bright and spin-polarized electron beams.

INTRODUCTION

Cutting-edge electron accelerator applications, such as X-ray Free Electron Lasers (XFELs), Ultrafast Electron Diffraction (UED) and Microscopy (UEM), as well as particle colliders including the Electron-Ion Collider (EIC), require high-quality electron beams. The quality of electron beams is characterized by their brightness. To enhance beam quality, it is essential to understand the factors that limit brightness. For high-charge-density applications, one key limitation of the 4D brightness B_{4D} is the accelerating electric field $\mathscr{E}$ at the photocathode surface, which determines the maximum charge density that can be extracted without degrading brightness

$$B_{4D} \propto \frac{\mathscr{E}^n}{MTE}, \tag{1}$$

where n varies between 1 and 2, depending on the photoinjector design. Another critical factor is the mean transverse energy (MTE) of the photoemitted electrons. MTE is defined as the average kinetic energy of emitted electrons in the direction perpendicular to the beam propagation. Due to different effects, the MTE of electrons emitted from conventional photocathodes is limited to several 100s of meV, which is nearly an order of magnitude larger than the theoretically predicted MTE limit ($MTE_{\min} \approx k_B T \approx 25$ meV at room temperature, where k_B is the Boltzmann constant and T is the temperature). Typical factors that limit MTE include the requirements for high charge density, the disordered nature of photocathode materials, surface roughness, and work function variations.

For high-energy-physics and nuclear-physics experiments that rely on spin-polarized electron sources, it is important to have very high electron spin polarization (ESP), which is defined as the degree to which the spin is aligned with a given direction and is given by

$$ESP = \left|\frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}\right|, \tag{2}$$

where $N_\uparrow$ and $N_\downarrow$ is the number of electrons with spins parallel and antiparallel to the direction of beam propagation, respectively. The ability of photocathodes to generate spin-polarized electron beams stems from the unique band structure of the photocathode materials. The maximum achievable ESP in experiments is determined by fundamental scattering mechanisms within these materials.

The overall quality of a photocathode is described by its quantum efficiency (QE), which is defined as the ratio of the number of emitted electrons N_{e^-} to the number of incident photons $N_{\hbar\omega}$

$$QE = \frac{N_{e^-}}{N_{\hbar\omega}}. \tag{3}$$

Photocathodes with higher QEs can produce electron beams with higher intensities.

Many applications require photocathodes with a prompt response time, which refers to the time it takes for a photocathode to emit electrons in response to incident light. The response time depends on the material's electron escape time, which is the time required for photoexcited electrons to reach the surface and escape into the vacuum.

It is also important to have a robust photocathode with long operational time, which means that photocathodes must operate under realistic photoinjector conditions. The photocathode reliability and stability under high electric-field loads and large laser fluences are crucial for sustained experimental operations.

Evidently, the quality of the electron beam is directly limited by the quality of the photocathode. Detailed theoretical photoemission modeling, combined with advanced photocathode characterization techniques, can be utilized to improve the performance of traditional electron sources and develop new ones with enhanced capabilities. Here, we will review some theoretical and experimental approaches towards generating high-quality electron beams for bright and spin-polarized applications.

PHOTOCATHODES FOR BRIGHT-BEAM APPLICATIONS

Traditionally, many accelerator facilities use metallic photocathodes to generate electron beams. Operating metallic photocathodes with photoexcitation energy close to the photoemission threshold results in low MTE but also yields low QE. While QE can be enhanced by increasing the photoexcitation energy, this also leads to an increase in MTE. For high-current, high-brightness applications, photocathodes can, in principle, operate near the photoemission threshold under high laser fluences. However, recent theoretical [1] and experimental [2] studies have shown that non-linear processes, such as multi-photon absorption, limit the MTE of electrons photoemitted from Cu to several hundred meV.

* chubenko@niu.edu

ISBN: 978-3-95450-219-6 ISSN: 2226-0366 doi: 10.18429/JACoW-LINAC2024-MOZA002

Due to the suppressed multi-photon absorption and reduced electron-electron interactions, low-electron-affinity semiconducting photocathodes may be better candidates to operate under high laser fluences. Indeed, our preliminary Monte Carlo calculations demonstrate (see Fig. 1) that under the photoinjector-meaningful laser power density the MTE of electrons photoemitted from thin GaAs activated to low electron affinity level is limited to several 10s of meV, which is an order of magnitude lower than that obtained for metals.

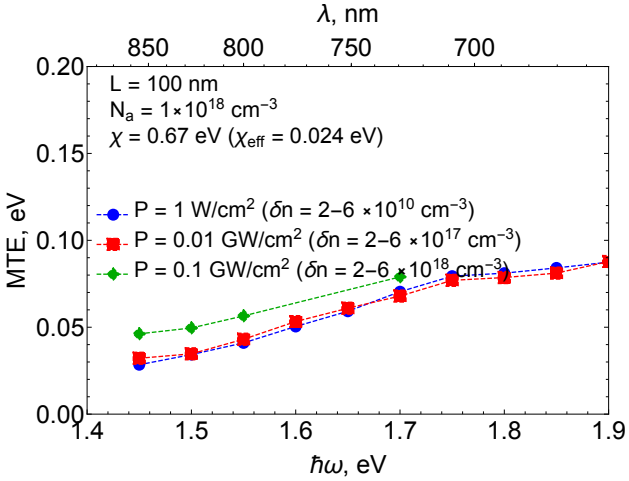

Figure 1: Preliminary Monte Carlo results for the MTE as a function of photoexcitation energy for different laser power densities from thin p-type GaAs activated to low electron affinity level.

Another class of low-electron-affinity photocathode materials is alkali antimonide films. These materials are being actively explored for generating extremely bright electron beams for XFELs as well as UED and UEM experiments. Cesium-antimonide photocathodes are the easiest alkali-antimonide materials to grow because they need only two particle evaporation sources. They can be easily deposited on substrates is a form of thin films. The substrate can enhance light absorption and act as a potential barrier preventing photoexcited electrons from diffusion into the bulk improving the photocathode response time. Cesium antimonides photoemit in a visible wavelengths range. They demonstrate thermal-limit MTE (≈ 30 meV) and relatively high QE (≈ 0.01 %) at the photoemission threshold (≈ 1.7 eV or 730 nm) [3, 4].

In general, these materials demonstrate rough surfaces when conventionally grown on rough disordered substrates. Surface roughness and work function variation are among the factors, which can limit MTE. It has been recently shown [5] that the cesium antimonide films grown on a lattice-matched single crystal strontium titanate substrate have ultra-low physical and chemical roughness, contributing less than 10 meV to MTE, even under high applied fields.

It has also been demonstrated [6] that Cs_3Sb films can be epitaxially grown on lattice-matched single-crystal SiC substrates. Epitaxial growth ensures that these materials have an ordered structure, resulting in atomically smooth surfaces and low MTE. Additionally, single-crystal Cs_3Sb exhibits higher QE in the red spectrum compared to disordered polycrystalline cesium antimonide [6].

It has been demonstrated that two phases of cesium antimonide, Cs_3Sb and CsSb, can be grown [7]. While CsSb exhibits a QE nearly an order of magnitude lower than that of Cs_3Sb, it shows up to 15 times greater robustness against oxidation. Therefore, this material is of particular interest for applications where ultra-high vacuum (UHV) conditions are not feasible. Further studies of these materials are required to identify factors limiting QE and affecting robustness. Additionally, recent experimental studies have demonstrated that electrons suffer significant energy losses before being emitted from thin film Cs_3Sb [3]. This phenomenon is not well understood, as electron-phonon interactions are expected to be suppressed in thin film materials. We use the Monte Carlo approach [8, 9] to model photoemission from Cs-Sb-compound materials to explain these phenomena and their effects on the photoemissive properties of these materials.

So, alkali-antimonide photocathodes demonstrate low MTE, relatively high QE (as compared to metals), and they can be deposited in a form of thin films resulting in a prompt photocathode response time. However, what about their robustness and operational lifetime under realistic photoinjector conditions? While there have been numerous tests of these materials in DC guns, there are only a few studies involving bialkali-antimonides in high-field RF/SRF guns, and no tests have been conducted on cesium antimonide photocathodes in RF/SRF environments.

The primary challenge in testing alkali antimonide photocathodes in accelerators is their sensitivity to vacuum conditions. These materials oxidize in air and must, therefore, be operated and transferred in an ultra-high vacuum (UHV) environment. Another issue is the incompatibility between photocathode growth systems and photoinjectors. Many university labs use flag-style sample holders for growing photoemitting films (Fig. 2). Fermi National Accelerator Laboratory (Fermilab) and some other institutions use INFN-style plugs. The Argonne Wakefield Accelerator (AWA) facility at Argonne National Laboratory (ANL) has three beamlines, each with its own unique photocathode plug style.

Northern Illinois University (NIU) is conveniently situated near two national laboratories with electron accelerator capabilities (Fermilab and ANL), making it an excellent location for developing a photocathode research and development program. NIU's Photoemission Research Laboratory (PRL, Fig. 3) is currently under development and already houses a thermal evaporation system previously used to grow cesium telluride photocathodes at Fermilab. This system, which utilizes an older INFN-type photocathode plug, will be upgraded with long-lasting effusion cells and a Reflection High-Energy Electron Diffraction (RHEED) system for *in situ/operando* characterization, aimed at achieving epitaxial growth of cesium antimonide photocathodes. The plug will also be modified to enable the growth of photoemitting films on semiconductor substrates. Additionally, a vacuum suitcase will be developed to enable transportation of photocathodes to accelerator facilities under UHV.

ISBN: 978-3-95450-219-6 ISSN: 2226-0366 doi: 10.18429/JACoW-LINAC2024-MOZA002

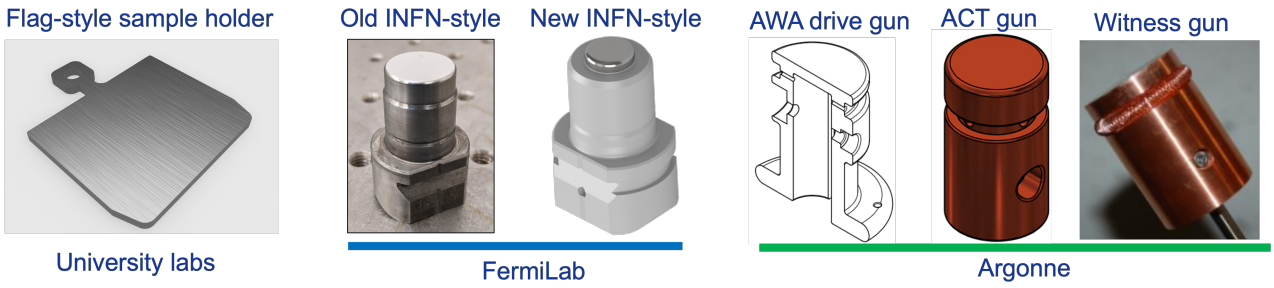

Figure 2: Examples of photocathode plugs used at university and national laboratories.

The work is also in progress to enable growth of alkali-antimonide photocathodes using the Pulsed Laser Deposition (PLD). Recent studies demonstrate that that this growth technique can be used to achieve atomically smooth ultra-thin (<30 nm) Cs_3Sb films with a high QE (>10%) [10].

The photocathodes grown at NIU will be tested at the Argonne Cathode Teststand (ACT), an L-band 1.3 GHz single-cell RF photocathode gun with emittance measurement capabilities. Currently, the ACT is only suitable for testing air-stable materials, but it will be upgraded to accommodate NIU-compatible photocathode plugs, allowing for the testing of photocathodes grown on semiconducting substrates. Additionally, a NIU-compatible load-lock system will be developed, and the pumping system will be updated to achieve a base pressure of $\sim 10^{-10}$ Torr. A deflecting cavity will also be added to enable photocathode response time measurements.

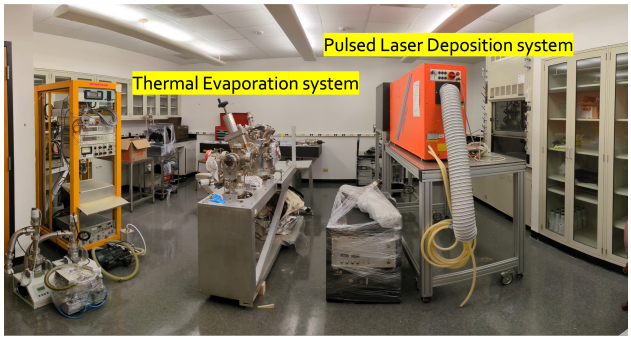

Figure 3: NIU's Photoemission Research Laboratory.

PHOTOCATHODES FOR SPIN-POLARIZED APPLICATIONS

Nowadays, spin-polarized electron beams for all applications are exclusively generated through the photoemission from GaAs-based photocathodes. Ability of GaAs photocathodes emit spin polarized electrons can be explained [8] by its unique band structure. GaAs has a strong spin-orbit coupling resulting in a significant energy gap between the split-off (so) and heavy-holes (hh)/light-holes (lh) subbands. When unstrained GaAs is illuminated by a circularly polarized light with the photon energy equal to the band-gap energy, only the transitions from hh subband into spin-up state and the transitions from the lh subband into spin-down state of the conduction band are possible. Probability of transition from hh band is 3 times higher than the probability of transition from lh. So, the maximum possible initial polarization is equal to 50%

$$ESP_0 = \left|\frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow}\right| = \left|\frac{3-1}{3+1}\right| = 50\%. \quad (4)$$

With increasing photon energy, the states of the so sub-band begin to be mixed in with the states of the lh and hh sub-bands. Because of this, the relative strength for the photoexcitation of electrons into different spin states of the conduction band changes and the degree of the initial spin polarization decreases from 50% to approximately 45% and then rapidly drops down when transitions from so sub-band become possible.

So, the initial electron spin polarization ESP_0 in unstrained GaAs is limited to 50%. However, due to different scattering mechanisms within the material, the experimental ESP is limited to $\sim$ 30%. Moreover, traditional methods used to increase QE lead to spin depolarization and vice versa. To explain the anti-correlation between QE and ESP, the spin-polarized Monte Carlo model of photoemission from bulk GaAs was developed [8]. The model accounts for both momentum-relaxation and spin-relaxation mechanisms. Therefore, both QE and ESP can be calculated simultaneously as a function of photon energy, doping density, and electron affinity level. The model provides good agreement with available experimental data in a wide range of photoexcitation energies and doping densities and therefore establishes a paradigm for future studies of spin-polarized photoemission.

Temperature Effects on Spin-Polarized Photoemission from Bulk GaAs

As it was mentioned above, the ESP of electrons extracted from unstrained GaAs is usually considerably less than the theoretical maximum of 50%. However, experimental observations have shown that ESP can be increased, and even exceed the theoretical 50% limit, at 77 K [11]. This phenomenon is not well understood, and it remains unclear what happens when the temperature is further reduced. To address these questions, ongoing work is using the Monte Carlo approach to study temperature effects on spin-polarized photoemission from bulk GaAs [12]. This approach relies on material band structure parameters, which can be obtained from Density Functional Theory (DFT). To validate the DFT

ISBN: 978-3-95450-219-6 ISSN: 2226-0366 doi: 10.18429/JACoW-LINAC2024-MOZA002

calculations, we first model electron transport at various temperatures and compare the results to experimental measurements.

Spin-Polarized Photoemission from Materials with Inherently Low/Negative Electron Affinity Levels

Future experiments at the EIC will require QE as high as several percent and ESP at least 85%. In principle, such high characteristics can be obtained from GaAs-based superlattice (SL) structures [13]. However, to achieve high QE, these photocathodes require Cs-based activation to negative electron affinity (NEA) to reduce the material work function. Unfortunately, this activation layer is very sensitive to vacuum conditions and gets destroyed due to the backward ion bombardment. Therefore, the QE degrades after several hours of operation. The alternative activation methods demonstrate the improved photocathode lifetime, but at the cost of QE [14].

We use the Monte Carlo approach to predict spin-polarized photoemission from other known spin-polarized materials and/or structures, which do not require traditional surface preparation. For example, polarization band engineering can be used to achieve an effective NEA condition without the use of Cs at the surface of GaN photocathode structures [15]. The work in progress to use the Monte Carlo approach in conjunction with DFT to study spin-polarized photoemission from III-Nitride materials.

Spin-Polarized Photoemission from CdTe and Other II-VI Semiconductors

Highly efficient GaAs-based SL structures are fabricated using an expensive molecular-beam epitaxy (MBE). Moreover, a limited number of vendors can provide those structures. One class of materials, which could be of potential interest for spin-polarized applications, is II-VI semiconductors including CdTe. They are of particular interest because they can be grown via cost-effective Atomic Layer Deposition (ALD) [16]. Moreover, CdTe demonstrates other properties desirable for highly efficient spin-polarized electron sources (see Table 1). For example, CdTe is a semiconductor material with a direct bandgap, which means it can absorb photons effectively and generate electron-hole pairs when illuminated with circularly polarized light. The degree of spin-polarization in a material often depends on the strength of its spin-orbit coupling. Materials with strong spin-orbit coupling tend to be more efficient in generating spin-polarized electrons when illuminated with circularly polarized light. CdTe has a moderate spin-orbit coupling. The surface quality of the CdTe material and the interface between CdTe and any potential coating or materials used in the device are crucial. A clean and well-prepared surface is essential to maximize the polarization of emitted electrons. The ALD technique can provide high-quality surfaces. Moreover, it can enable p-type doping required for achieving high QE. Therefore, the Monte Carlo approach combined with DFT band-structure calculations is in progress to study spin-polarized photoemission from CdTe and other II-VI semiconductors

Table 1: Comparison of GaAs and CdTe for spin-polarized photocathode applications. The color coding indicates desirable characteristics (green), less desirable characteristics (yellow), and undesirable characteristics (red).

	GaAs	**CdTe**
Direct bandgap	Yes	Yes
Cs-based activation	Yes	Yes
Surface quality	High	High
p-doped	Yes	Yes
Spin-orbit coupling	Strong	Moderate
Cost	MBE, expensive	ALD, cheap
Accessibility	Limited	Accessible

Spin-Polarized Photoemission from Alkali-Antimonide-Based Structures

Recent studies indicate that alkali-antimonide-based photocathodes such as Na_2KSb/Cs_3Sb heterostructures can be also used to generate spin-polarized electron beams [17]. Taking into account the fact that (bi)alkali antimonides are more robust to vacuum conditions as compared to cesiated GaAs ($\sim 10^{-10}$ Torr vs. $<10^{-11}$ Torr) and demonstrate low electron affinity levels without need of activation process, these materials can potentially outperform and replace cesiated GaAs to produce intense highly spin-polarized electron sources. The work is in progress to model material and photoemission properties necessary to identify the specific material properties required for achieving high QE and high ESP from these materials.

CONCLUSION

Alkali-antimonide photocathodes are promising candidates for generating ultra-bright electron beams for advanced accelerator applications. Ultra-smooth, low-MTE alkali-antimonide films can be produced via MBE on lattice-matched semiconducting substrates, using *in situ* characterization with RHEED. Ongoing work aims to enable the deposition of high-quality single-crystal alkali-antimonide photocathodes on photocathode plugs compatible with the ACT beamline and further testing of these photocathodes under high electric fields and large laser fluences.

Although GaAs-based photocathodes can generate high-QE, high-ESP electron beams, research and development of alternative spin-polarized electron sources that could outperform and potentially replace GaAs technology is highly encouraged. Ongoing efforts are utilizing the Monte Carlo approach in combination with DFT calculations to investigate novel spin-polarized materials.

ACKNOWLEDGEMENTS

The author thanks the Center for Bright Beams, NSF award PHY-1549132.

ISBN: 978-3-95450-219-6 ISSN: 2226-0366 doi: 10.18429/JACoW-LINAC2024-MOZA002

REFERENCES

[1] 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. 244 903, 2018. doi:10.1063/1.5053082

[2] C. J. Knill *et al.*, "Effects of nonlinear photoemission on mean transverse energy from metal photocathodes", *Phys. Rev. Accel. Beams*, vol. 26, no. 9, p. 093 401, 2023. doi:10.1103/PhysRevAccelBeams.26.093401

[3] A. Kachwala, P. Saha, P. Bhattacharyya, E. Montgomery, O. Chubenko, and S. Karkare, "Demonstration of thermal limit mean transverse energy from cesium antimonide photocathodes", *Appl. Phys. Lett.*, vol. 123, no. 4, p. 044 106, 2023. doi:10.1063/5.0159924

[4] E. A. Taft and H. R. Philipp, "Structure in the energy distribution of photoelectrons from K_3Sb and Cs_3Sb", *Phys. Rev.*, vol. 115, no. 6, p. 1583, 1959. doi:10.1103/PhysRev.115.1583

[5] P. Saha *et al.*, "Physically and chemically smooth cesium-antimonide photocathodes on single crystal strontium titanate substrates", *Appl. Phys. Lett.*, vol. 120, no. 19, p. 194 102, 2022. doi:10.1063/5.0088306

[6] C. Parzyck *et al.*, "Single-crystal alkali antimonide photocathodes: High efficiency in the ultrathin limit", *Phys. Rev. Lett.*, vol. 128, no. 11, p. 114 801, 2022. doi:10.1103/PhysRevLett.128.114801

[7] C. Parzyck *et al.*, "Atomically smooth films of cssb: A chemically robust visible light photocathode", *APL Mater.*, vol. 11, no. 10, p. 101 125, 2023. doi:10.1063/5.0166334

[8] O. Chubenko *et al.*, "Monte Carlo modeling of spin-polarized photoemission from p-doped bulk GaAs", *J. Appl. Phys.*, vol. 130, p. 063 101, 2021. doi:10.1063/5.0060151

[9] S. Karkare *et al.*, "Monte Carlo charge transport and photoemission from negative electron affinity GaAs photocathodes", *J. Appl. Phys.*, vol. 113, no. 10, p. 104 904, 2013. doi:10.1063/1.4794822

[10] W. Stam *et al.*, "Growth of ultra-flat ultra-thin alkali antimonide photocathode films", *APL Mater.*, vol. 12, no. 6, p. 061 114, 2024. doi:10.1063/5.0213461

[11] W. Liu, M. Poelker, X. Peng, S. Zhang, and M. Stutzman, "A comprehensive evaluation of factors that influence the spin polarization of electrons emitted from bulk gaas photocathodes", *J. Appl. Phys.*, vol. 122, no. 3, p. 035 703, 2017. doi:10.1063/1.4994306

[12] J. Callahan, O. Chubenko, L. Cultera, S. Karkare, A. Rodriguez Alicea, and J. O. Mendez-Nieves, "Monte carlo modeling of spin-polarized photoemission from gaas with low-temperature and strained-lattice effects", in *Proc. 15th International Particle Accelerator Conference*, Nashville, TN, pp. 2119–2122, 2024. doi:10.18429/JACoW-IPAC2024-WEPC65

[13] W. Liu *et al.*, "Record-level quantum efficiency from a high polarization strained gaas/gaasp superlattice photocathode with distributed bragg reflector", *Appl. Phys. Lett.*, vol. 109, no. 25, p. 252 104, 2016. doi:10.1063/1.4972180

[14] J. K. Bae, A. Galdi, L. Cultrera, F. Ikponmwen, J. Maxson, and I. Bazarov, "Improved lifetime of a high spin polarization superlattice photocathode", *J. Appl. Phys.*, vol. 127, no. 12, p. 124 901, 2020. doi:10.1063/1.5139674

[15] J. Marini, I. Mahaboob, E. Rocco, L. Bell, and F. Shahedipour-Sandvik, "Polarization engineered n-polar cs-free gan photocathodes", *J. Appl. Phys.*, vol. 124, no. 11, p. 113 101, 2018. doi:10.1063/1.5029975

[16] H. Bhandari, "Cost-effective atomic layer deposition growth of chalcogenide-based polarized electron sources (cdte, znte, mnte, etc.)", in *Proc. Photocathode Physics for Photoinjectors (P3) Workshop*, Stony Brook, NY, 2023. https://indico.bnl.gov/event/19730/.

[17] V. Rusetsky *et al.*, "New spin-polarized electron source based on alkali antimonide photocathode", *Phys. Rev. Lett.*, vol. 129, no. 16, p. 166 802, 2022. doi:10.1103/PhysRevLett.129.166802