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MONTE CARLO STUDY OF ELECTRON ENERGY LOSSES AND STOICHIOMETRY EFFECTS IN THIN CESIUM ANTIMONIDE PHOTOCATHODES*

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Abstract

Cesium antimonide photocathodes are known for their ability to generate bright electron beams for various accelerator applications. Lab-grown polycrystalline cesium antimonides as well as Cs_1Sb and Cs_3Sb crystals are distinguishable; however, it remains unclear how the crystalline and other material properties of each govern the main photocathode properties such as quantum efficiency and mean transverse energy. Furthermore, photoexcited electrons undergo significant energy losses before being emitted from thin cesium antimonide films. This process is not well understood since there is very little room for scattering events within thin films. The generation of ultra-bright electron beams, capable of substantially enhancing the scientific potential of advanced accelerator applications, requires deep understanding of these and other fundamental mechanisms, which constrain photocathode performance and simultaneously determine the maximum attainable beam brightness. The purpose of this work is to use the Monte Carlo approach in a combination with Density Functional Theory to shed light on these mechanisms and provide the guidance for effective photocathode optimization.

INTRODUCTION

High-quality beams are required for operating x-ray free electron lasers, ultrafast electron diffraction experiments, particle colliders, and other advanced accelerator applications. Such beams are produced by photocathodes primarily characterized by high Quantum Efficiency (QE) and low Mean Transverse Energy (MTE) defined as

$$QE = \frac{\#\ of\ emitted\ electrons}{\#\ of\ incident\ photons}, \tag{1}$$

$$MTE = \frac{m < v_\perp^2 >}{2}, \tag{2}$$

where m is the electron's mass, $v_\perp$ is the velocity of emitted electrons in the direction perpendicular to beam propagation.

The QE and MTE are important qualities of photocathodes as they characterize how bright the beam they produce will be. A larger QE indicates that more electrons per incident photon are emitted and are thus favorable for efficient beams. A low MTE is the result of electrons emitting perpendicularly to the flat surface of the photocathode. This ensures that the electrons maintain a more cohesive trajectory in the beam. The beam brightness B is related to MTE through the following expression [1]

$$B \propto \frac{E^n}{MTE}, \tag{3}$$

where E is the electric field applied at the photocathode surface and n is a real number between 1 and 2 as defined by the photoinjector design.

Because of their near-thermal-limit MTE and relatively high QE compared to metals, cesium antimonide photocathodes are promising candidates for high-brightness applications [2]. Effective optimization of these photocathodes requires deep understanding of fundamental processes governing photoemission process. For example, emitted electrons from bulk crystals experience a drop in energy after emission which is attributed to energy loss from collisions during the transport phase. It was thought that since thin-films have a shorter transport space, they would experience a less significant energy loss. However, recent experimental data [2] shows that thin-film energy loss is comparable to that of bulk crystals. A more complete understanding of this process in Cs-Sb compound photocathodes will help to enhance photocathode capabilities towards generating brighter electron beams. Moreover, recent experimental data obtained by Maxson's group at Cornell University demonstrates that Cs_1Sb photocathodes exhibit reduced QE but are less sensitive to vacuum conditions as compared to Cs_3Sb, which makes these materials attractive for applications where robustness is a key parameter.

The goal of this project is to utilize Monte Carlo simulations in conjunction with Density Functional Theory (DFT) to determine the effects of crystal stoichiometry and explain energy losses for electrons emitted from thin films.

THEORETICAL APPROACH

The earlier developed Monte Carlo model [3] of photoemission has been modified and used to calculate QE and MTE from Cs-Sb compound materials. The model incorporates three fundamental steps of photoemission (Fig. 1) and simulates the excitation of an electron from the valence band all the way to emission and recalculates the energy after every scattering event during the transport stage.

The Monte Carlo model requires knowledge of many band model parameters. Typically, those parameters can be obtained from experimental measurements. However, limited

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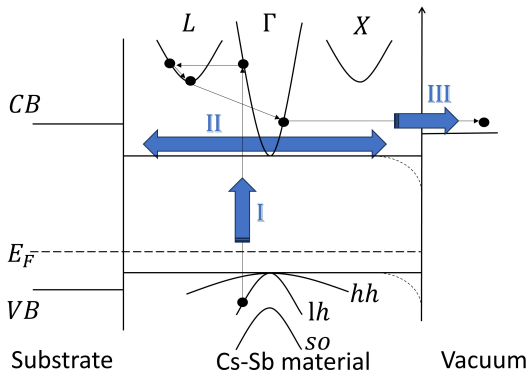

Figure 1: 3-Step model of photoemission from semiconductors: I - photoexcitation of electrons from the Valence Band (VB) to the Conduction Band (CB); II - transport of electrons to the surface; III - emission into the vacuum.

experimental data are available for Cs_3Sb or Cs_1Sb. Therefore, DFT band structure calculations should be employed. The complete list of preliminary obtained modeling parameters is outlined in Table 1.

PRELIMINARY RESULTS AND DISCUSSIONS

The electron loses energy as it scatters, but it may gain energy through the absorption of phonons. A net energy loss is expected as phonon emission is the dominant process as indicated in Fig. 2.

Our preliminary results for the emitted energy distribution (Fig. 3) are in good qualitative agreement with experimental data [2]. However, our model is predicting the emitted electron energy slightly higher than what is found in experimental data. Further study is ongoing to explain this discrepancy.

Other experimental data [4] shows that the absorption length is significantly higher for Cs_1Sb than Cs_3Sb in the low energy region (Fig. 4). This correlates with Cs_1Sb exhibiting a higher QE than Cs_3Sb in the low energy regime, where the MTE achieves its minimum values. In addition to allowing the crystals to be distinguished, this suggests that Cs_1Sb photocathodes have the potential to achieve more desirable efficiency than Cs_3Sb. The performance of both crystals at

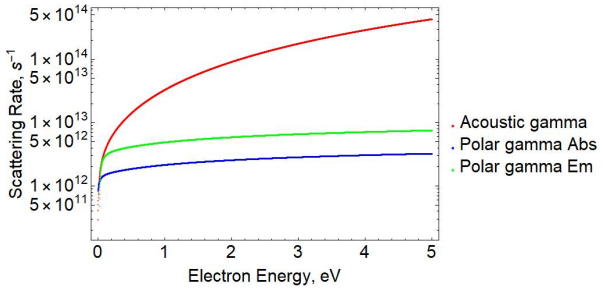

Figure 2: Absorption and emission scattering rates for the gamma valley of Cs_3Sb.

Table 1: Preliminary Obtained Model Parameters used in the Simulations

Parameter, units	Value
Energy bandgap, eV	1.6
Energy split-off, eV	0.47
Energy light/heavy hole, eV	0.0
Valley-splitting L-Γ, eV	1.4
Valley splitting X-Γ, eV	0.256
Number of equivalent Γ valleys	1.0
Number of equivalent L valleys	4.0
Number of equivalent X valleys	3.0
Γ valley relative electron mass, m_0	0.304
L valley relative electron mass, m_0	1.0
X valley relative electron mass, m_0	0.341
Density of states mass Γ, m_0	0.23
Density of states mass L, m_0	0.23
Density of states mass X, m_0	0.23
Heavy hole mass, m_0	10.472
Light hole mass, m_0	2.426
Split-off mass, m_0	3.174
Γ non-parabolicity, eV^{-1}	0.822
L non-parabolicity, eV^{-1}	0.0
X non-parabolicity, eV^{-1}	0.561
Heavy hole non-parabolicity, eV^{-1}	0.0
Light hole non-parabolicity, eV^{-1}	0.0
Split-off non-parabolicity, eV^{-1}	0.0
High frequency dielectric constant, ϵ_0	8.2
Static dielectric constant, ϵ_0	8.96
Crystal density, kg m^{-3}	4519
Sound velocity, m s^{-1}	5153
Γ acoustic deformation potential, eV	7.0
L acoustic deformation potential, eV	9.2
X acoustic deformation potential, eV	9.0
Polar optical phonon energy, eV	0.022
Γ-L valley deformation potential, eV $Å^{-1}$	10
Γ-X valley deformation potential, eV $Å^{-1}$	10
L-Γ valley deformation potential, eV $Å^{-1}$	10
L-L valley deformation potential, eV $Å^{-1}$	10
L-X valley deformation potential, eV $Å^{-1}$	5.0
X-Γ valley deformation potential, eV $Å^{-1}$	10
x-L valley deformation potential, eV $Å^{-1}$	5.0
X-X valley deformation potential, eV $Å^{-1}$	7.0
Γ-L intervalley phonon energy, eV	0.0278
Γ-X intervalley phonon energy, eV	0.0299
L-Γ intervalley phonon energy, eV	0.0278
L-L intervalley phonon energy, eV	0.029
L-X intervalley phonon energy, eV	0.0293
X-Γ intervalley phonon energy, eV	0.0299
X-L intervalley phonon energy, eV	0.0293
X-X intervalley phonon energy, eV	0.0299

incidence energies above 2.4 eV become comparable to each other.

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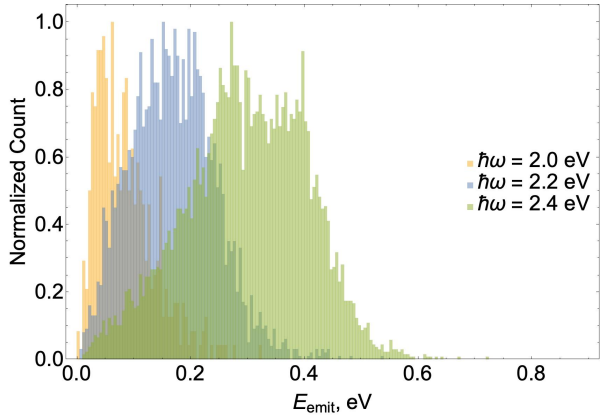

Figure 3: Energy distribution of emitted electrons predicted by Monte Carlo simulation.

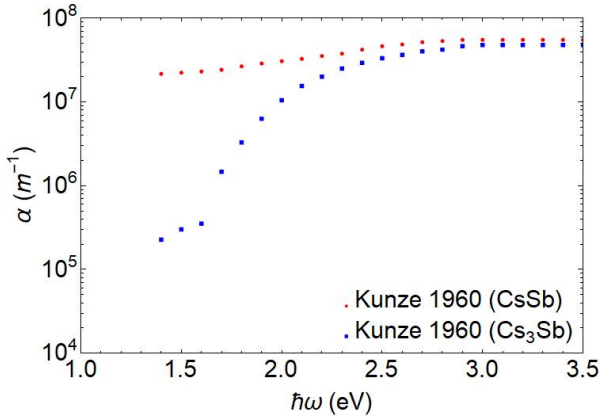

Figure 4: Experimental absorption coefficient for Cs_3Sb and Cs_1Sb [4].

CONCLUSIONS AND FUTURE FORK

The Monte Carlo model, in combination with DFT calculations, was used to calculate QE, MTE, and emission energy distributions comparable to existing experimental data. These calculations can be used to investigate various scattering mechanisms and their effect on the energy of emitted electrons. This is a promising method to begin to explain why electrons emitted from thin-films lose energy similarly to electrons emitted from bulk crystals.

Future work on the project is focused on improving the model's versatility and accounting for realistic conditions. The model is currently using parameters for Cs_3Sb but as more data for Cs_1Sb becomes available via DFT, the model will be able to do calculations for both crystals. There are experimental values for absorption length available for Cs_1Sb that are used to calculate the real and imaginary part of the dielectric function but these will be updated when possible. To improve model accuracy by accounting for realism, it will be updated to include defect scattering effects that occur in polycrystalline akali antimonide photocathodes.

As the model grows more accurate, it will be a more effective tool for studying energy loss in photocathode photoemission. Further use of the model and study of crystal effects on emitted energy distribution, QE, and MTE will ultimately lead to being able to grow photocathodes that produce better, brighter beams.

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