Novel Ultrabright Photocathodes Discovered from Machine Learning and Density Functional Theory Driven Screening

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Talk Overview

1.) Development of a generalizable photoemission model
https://doi.org/10.1103/PhysRevB.101.235447

2.) Machine learning for predicting the work function of photocathodes
(P. Schindler, E. R. Antoniuk et al., arXiv, 2020)
arXiv:2011.10905
Code:
https://github.com/peterschindler/WorkFunctionDatabase

3.) Screening for novel photocathode materials
(E. R. Antoniuk et al., Advanced Materials, 2021)
https://doi.org/10.1002/adma.202104081
Material databases enable computational materials discovery

# Synthesized Inorganic Materials Per Decade
(from ICSD)

- More materials synthesized in 2010s than all materials before 1990!
- We have \textit{tripled} our knowledge of known materials since 1990

![Graph showing the number of synthesized inorganic materials per decade with a significant increase in the 2010s.]
Computational materials discovery requires development of screening methodologies to be successful.

Grand Goal: Can we search through all known materials to find the most promising photocathode material?

~144,000 candidates

Required Properties

$$ Billion Dollar Photocathode Material $$
We develop new methods for predicting photocathode properties.

Properties We Have:
- Crystal Structure
- Electronic Band Structure
- Intrinsic Emittance

Properties We Need:
- Work Function
- Intrinsic Emittance
We Develop a Generalizable Photoemission Model

Conceptual Idea:
1.) Identify photoemitting states for $\hbar \omega$.
2.) Calculate photoemission probability for each pair of states.
3.) Calculate intrinsic emittance as weighted average of photoemitting states.
4.) Repeat for new $\hbar \omega$.

\[ \varepsilon_{int}(\omega, T) = \frac{\hbar}{mc} \sqrt{\sum_i w_i(\mathbf{k}, \omega, T) \cdot k_x^2}, \]
We Develop a Generalizable Photoemission Model

Calculate Emittance:

\[ \varepsilon_{int}(\omega, T) = \frac{\hbar}{mc} \sqrt{\sum_i w_i(\vec{k}, \omega, T) \cdot k_{x,i}^2} \]

Our Photoemission Model Generalizes to All Tested Materials

- DFT predictions quantitatively agree with experiments for all materials

Experimental Precision

We use machine learning to identify novel photocathode materials

1.) Calculate DFT work functions of 29,270 surfaces

2.) Machine learning:

\[ \phi_{DFT} = 3.2eV \]
\[ \phi_{DFT} = 5.7eV \]
\[ \vdots \]

Repeat ~1 million times

Code: https://github.com/peterschindler/WorkFunctionDatabase
We use machine learning to identify novel photocathode materials

- Test error (0.19eV) comparable to DFT!
- DFT: ~1 month for 20,000 materials
- ML: ~1 hour for 100,000 materials

Code: https://github.com/peterschindler/WorkFunctionDatabase

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3.) Screening for novel photocathode materials
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We screen for low emittance photocathode materials

Screening #1: Finding all possible low intrinsic emittance photocathodes

Screening principles:

i) Do fast steps first

ii) Minimize false negatives

iii) Validate everything after screening
We utilize our generalizable photoemission model to screen over 7,981 semiconducting materials from Materials Project. We screen 7,981 semiconducting materials from Materials Project. Minimize false negatives by assuming best possible work function. Antoniuk et al., Advanced Materials (2021)
We determine the emittance of 7,981 semiconducting materials.

Antoniuk et al., Advanced Materials (2021)
We identify photocathode materials that are robust against work function variances

We identify 1070 materials that are:

- Synthesizable
- Thermodynamically Stable
- Emit for photon energies between 1-5eV
- Insensitive to work function variance

Antoniuk et al., Advanced Materials (2021)
We identify photocathode materials that are commercially available.

We identify 13 commercially available photocathode materials.

A. Semiconducting Band Structure
B. Low Intrinsic Emittance Score
C. Synthesizable + Stable
D. Work Function Robust
E. Commercial Availability

Antoniuk et al., Advanced Materials (2021)
We Identify Commercially Available Photocathode Materials with Ultralow Intrinsic Emittance

We recalculate emittance with high accuracy method

11/13 materials achieve comparable emittance as $K_2$CsSb

Antoniuk et al., Advanced Materials (2021)
We Identify Air Stable Visible Light Photocathode Materials

Screening #2: Finding low emittance, air-stable, visible active photocathodes

We perform **multi-objective screening** to materials that are:

i) Air-stable  
ii) Visible light active  
iii) Low emittance
We use machine learning to identify all low emittance visible light photocathodes

2,114 materials → 90,698 surfaces

We identify all materials with any work function < 3eV surface

Antoniuk et al., Advanced Materials (2021)
We utilize our dataset to assess the predictive power of conventional wisdom.

Conventional wisdom: Alkali and alkaline-earth metals have low work functions.

- We analyze ML work functions of **90,698 surface slabs**

99.9% of <3eV work function materials come from here!

Antoniuk et al., Advanced Materials (2021)
We Identify M$_2$O as Air Stable Visible Light Photocathode Materials

**Air stability**: Look for **oxide** binaries

We find: **Na$_2$O, K$_2$O, Rb$_2$O**

Literature reports confirm air-resistance!

Antoniuk et al., Advanced Materials (2021)
We Identify $\text{M}_2\text{O}$ as Air Stable Visible Light Photocathode Materials

- Search list of visible light photocathodes for **air stable oxides**
- We discover $\text{M}_2\text{O}$ **family** of photocathode materials

Similar emittance, but **air stable**!

Potential air-stable, visible photocathodes!

Antoniuk et al., Advanced Materials (2021)
Collaborators
Dr. Peter Schindler (Stanford)
Prof. Piero Pianetta (SLAC)
Prof. W. Andreas Schroeder (UIC)
Dr. Bruce Dunham (SLAC)
Dr. Theo Vecchione (SLAC)
Prof. Evan Reed (Stanford)
Jaclyn Schillinger
<table>
<thead>
<tr>
<th>Known Photocathode Family</th>
<th>Materials In Family</th>
<th># Materials in Family</th>
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</thead>
<tbody>
<tr>
<td>Alkali Tellurides ((Cs_2Te))</td>
<td>Li_2Te (mp-2530), CsTe (mp-8361), K_2Te (mp-1747), Cs_2Te (mp-573763), KTe (mp-2072), Rb_2Te (mp-441), CsNaTe (mp-5339), Na_2Te (mp-2784), KLiTe (mp-4495), KNaTe (mp-8755), RbTe (mp-8360)</td>
<td>11</td>
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<tr>
<td>Alkali Antimonides ((Cs_3Sb, K_2CsSb))</td>
<td>Cs_3Sb (mp-10378), Li_3Sb (mp-2074), Rb_3Sb (mp-16319), RbSb (mp-10487), RbSb (mp-7444), K_3Sb (mp-10159), NaLi_2Sb (mp-5077), CsSb (mp-573514), CsK_2Sb (mp-581024), KSB (mp-1536), KNaSb (mp-15724)</td>
<td>11</td>
</tr>
<tr>
<td>III-V Semiconductors ((GaS, GaAs))</td>
<td>AlAs (mp-2172), AlN (mp-1700), GaN (mp-830), GaP (mp-2490), InN (mp-22205), BP (mp-1479), AlSb (mp-1018100), AlSb (mp-2624), GaSb (mp-1156), GaAs (mp-2534), GaSb (mp-1018059), AlGa_3N_4 (mp-1019508), AlP (mp-1550), InP (mp-966800)</td>
<td>14</td>
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