Ultra-bright Designer Photocathodes

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Outline

- Introduction
- Approach, results, and plans:
  - Design
  - Synthesize
  - Characterize
- Summary

Introduction

- Ultra-bright photocathodes are a key technology for the development of future light sources.
- Two approaches are applicable, depending on time-frame:
  - **Near-term**: Optimize the synthesis and performance of long-known photocathodes such as Cs_2Te, CsKSb, GaAs (ANL, ASTeC, BNL, HFZD, INFN, JLab, LBNL, PITZ, et al.)
  - **Mid-to-far-term**: Explore novel crystal systems numerically and optimize ("design") their properties, or nano-engineer surfaces, or other novel idea (**ANL**, ASTeC, BNL, Eindhoven, Jlab, LBNL, SLAC, UCLA, Vanderbilt, et al.).
- Properties to design or tune include:
    - Intrinisic emittance
    - Workfunction
    - QE
    - Reliability, robustness (vacuum, E-field, field emission)

Approach to cathode design

- **Design** *(accelerator & condensed matter physics, computational chemisty)*
  - Derive cathode requirements from future light source requirements.
  - Investigate candidate systems; compute electronic properties.
  - Select systems with promising predicted properties for synthesis.
- **Synthesize** *(chemistry, materials science)*
  - Develop & test hypotheses for chemical reactions.
- **Characterize** *(surface science, materials science)*
  - Develop portable load-lock system.
  - Develop cross-compatible sample transfer system.
  - Compare measurements with predictions.

Design

Main tool:

- Quantum-chemical Density Functional Theory (DFT)-based surface-physics calculations.

Two codes used:

- PWSCF (crystal lattice parameters, workfunction, gap energy, surface energy)
- YAMBO (optical absorption spectra)

Two of our cathode concepts:

- Tuning workfunction and intrinsic emittance using ultrathin films.
- Tuning the workfunction of Cs_2Te.

Potential low-transverse-emittance layered structure

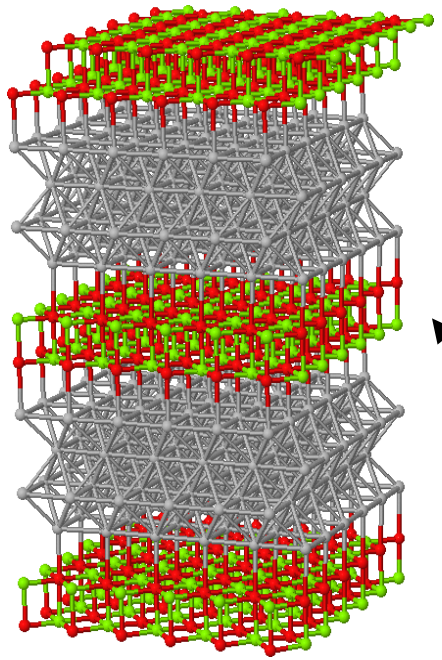
Preliminary idea from surface catalysis systems [L. Giordano et al., J. Chem. Phys. 127, 144713 (2007)].

k_x vs. energy bands relative to E_f

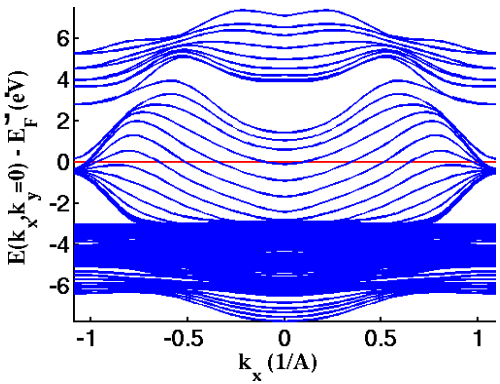
Lowest-energy (relative to E_f) surface bands in k_x vs. k_y space

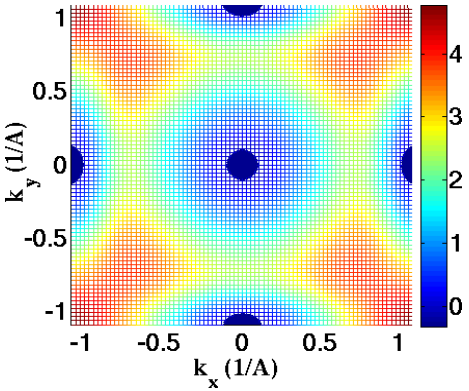
K. Nemeth et al., PRL 104, 046801 (2010).

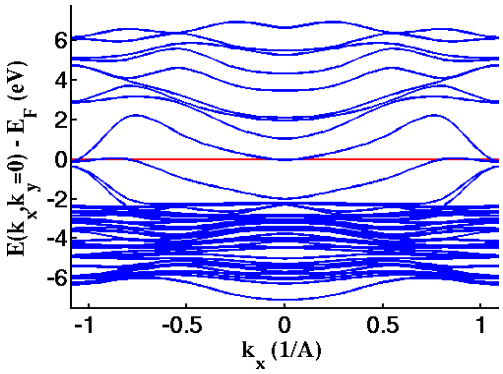
Ag(001)16L
DFT(PBE)
k_{max} 0.1 Å^{-1}

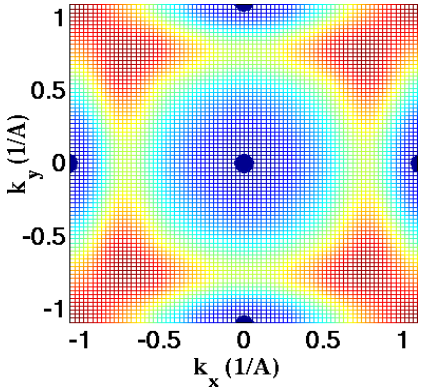


k_{max}
0.05 Å^{-1}





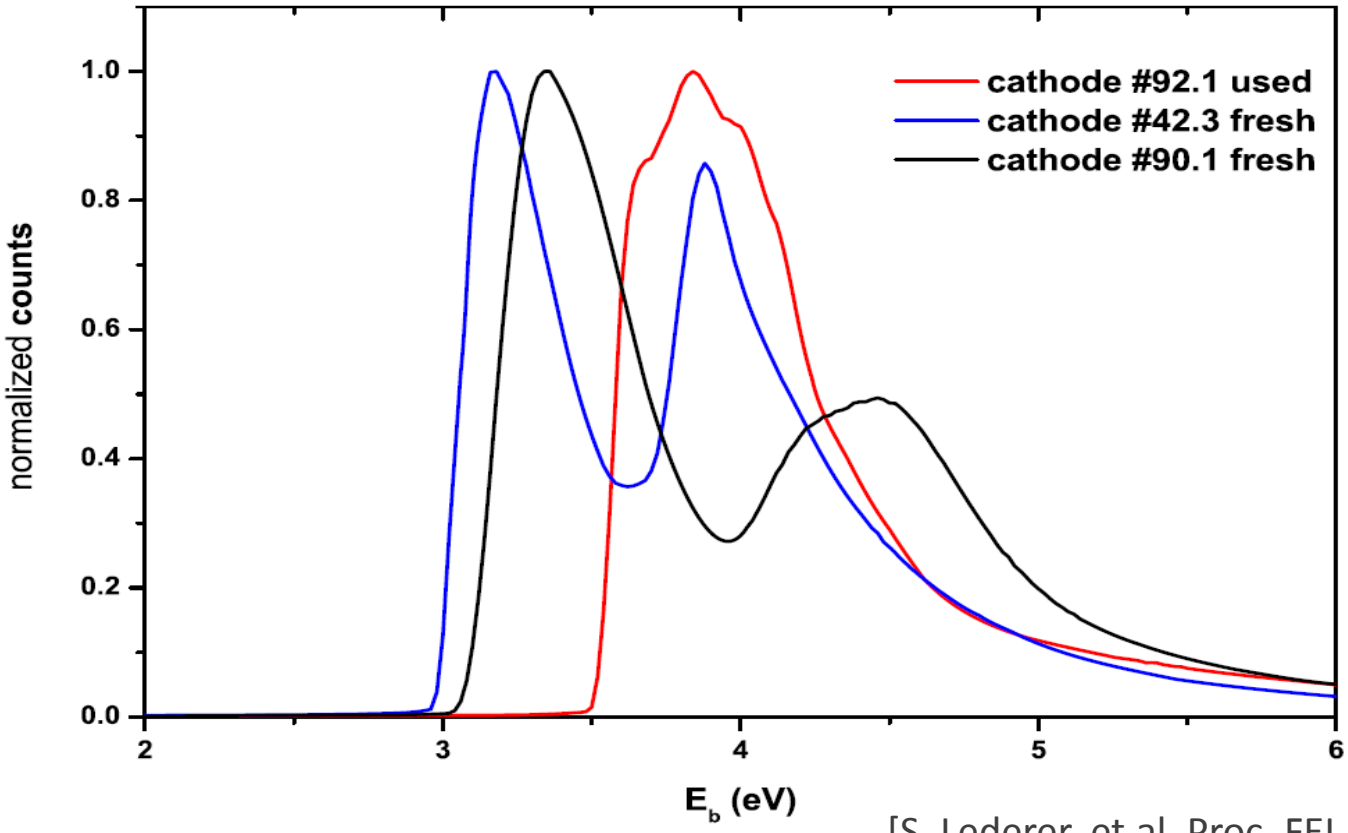




MgO(100)2L-Ag(100)4L-MgO(100)2L; DFT(PW91)
Work function reduced by ~1.6 eV relative to pure Ag(001)

$\varepsilon_{n,i}/\sigma_x$ 0.18 mm-mr/mm $E_{thermal}$ 30 meV

Photoemission from Cs_2Te cathode



[S. Lederer, et.al, Proc. FEL 2007, p. 457]

- Workfunction increases and QE decreases as Cs_2Te ages.
- Pushing the Cs_2Te photoemission (workfunction) to visible/infrared would have tremendous advantages!

Quantum efficiency of Cs_2Te exposed to small gas molecules, such as O_2, CO_2, CO, N_2 and CH_4

[A. di Bona, et.al. J.Appl.Phys. 80 (5), 1996]

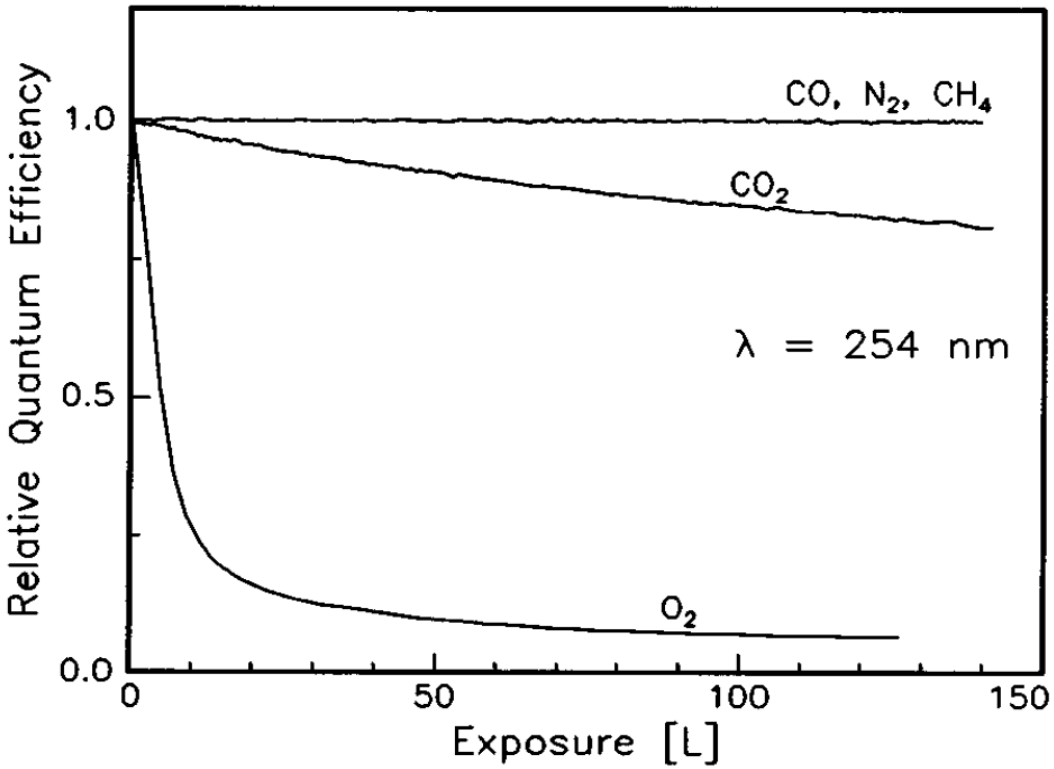


FIG. 7. Quantum efficiency drop for the 254 nm radiation of the photocathode exposed to different gases.

- Another important, relatively common small molecule, **acetylene, C_2H_2** (e.g. for welding), was not investigated.
- **What about reacting Cs_2Te with acetylene?**
- Acetylene is fairly reactive, easily losing its hydrogens, which takes electrons away from the material in contact. This forms "acetylides," containing the acetylide anion $[:C:::C:]^{2-}$, commonly denoted as C_2^{2-}.

A_2MC_2 Type Ternary Acetylides

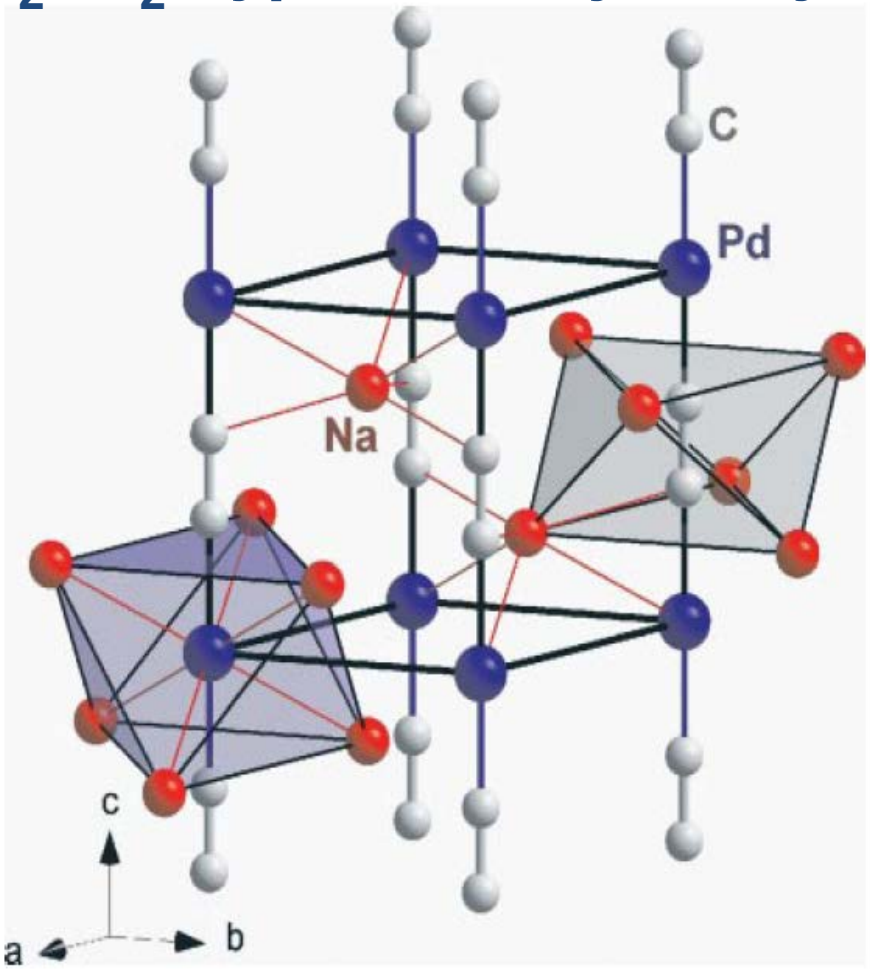


Fig. 3 Crystal structure of Na_2PdC_2 ($P\bar{3}m1$, Z = 1). Selected interatomic distances are: Pd−C 200.2(2) pm, Na−Pd 296.8(2) pm, Na−C 263.1(1) pm, C−C 126.3(3) pm.

[Fig: U. Ruschewitz, Z. Anorg. Allg. Chem. 632, 705, 2006.]

- Unique 1D substructures, where A=alkali metal and M=transition metal or metalloid.
- All known A_2MC_2 are colored semiconductors with 2.1-2.8 eV direct bandgaps.
- Synthesized so far: M∈[Pt,Pd] and A∈[Na,K,Rb,Cs].
- **We investigated acetylated Cs_2Te, where M=Te and A=Cs**
- Completely new class of materials for photophysics and low-workfunction applications (e.g., field-emission displays). *Provisional patent application filed: K. Nemeth et al.*

Experimental vs. DFT-calculated workfunctions (ϕ)

TABLE III: Experimental and calculated (DFT) properties of photoemissive surfaces of validation materials: workfunctions (Φ), bandgaps at the Γ-point $E_g(\Gamma)$ and surface energies (σ).

Compound and surface	Φ (eV) EXPT	Φ (eV) DFT	$E_g(\Gamma)$ (eV) DFT	σ (eV/Å^2) DFT
Cs(100)	2.14 [21]	2.00	0.29	0.005
Te(001)	4.95 [21]	5.02	0.54	0.036
$Cs_2Te(001)$	2.90-3.0 [22]	3.08	0.77	0.015
$Cs_2Te(010)$	2.90-3.0 [22]	2.90	1.04	0.014
$(Cs)Na_3KSb$	1.55 [23]	-	-	-
K_2CsSb	1.9-2.1 [24, 25]	-	-	-

J.Z. Terdik, K. Nemeth, K. Harkay, et al, submitted for publication.

- Good agreement for Cs, Te, Cs_2Te; we are confident of predictions for theoretical systems with these components.
- Note low workfunctions for K_2CsSb and $(Cs)Na_2KSb$ (engineered for high QE).

Calculated ϕ for selected A_2MC_2

- Significant ϕ anisotropy among rod-perpendicular (001) and rod-parallel, (110) and (010), orientations.
- Cs_2TeC_2 (010) and Cs_2PdC_2 (010) are photo-emissive in visible to near-IR!
- Comparable-to-better ϕ than K_2CsSb and (Cs)Na_2KSb (1.55-2.1 eV).
- Rod-perpendicular surface could act like a 2D nanotip array.

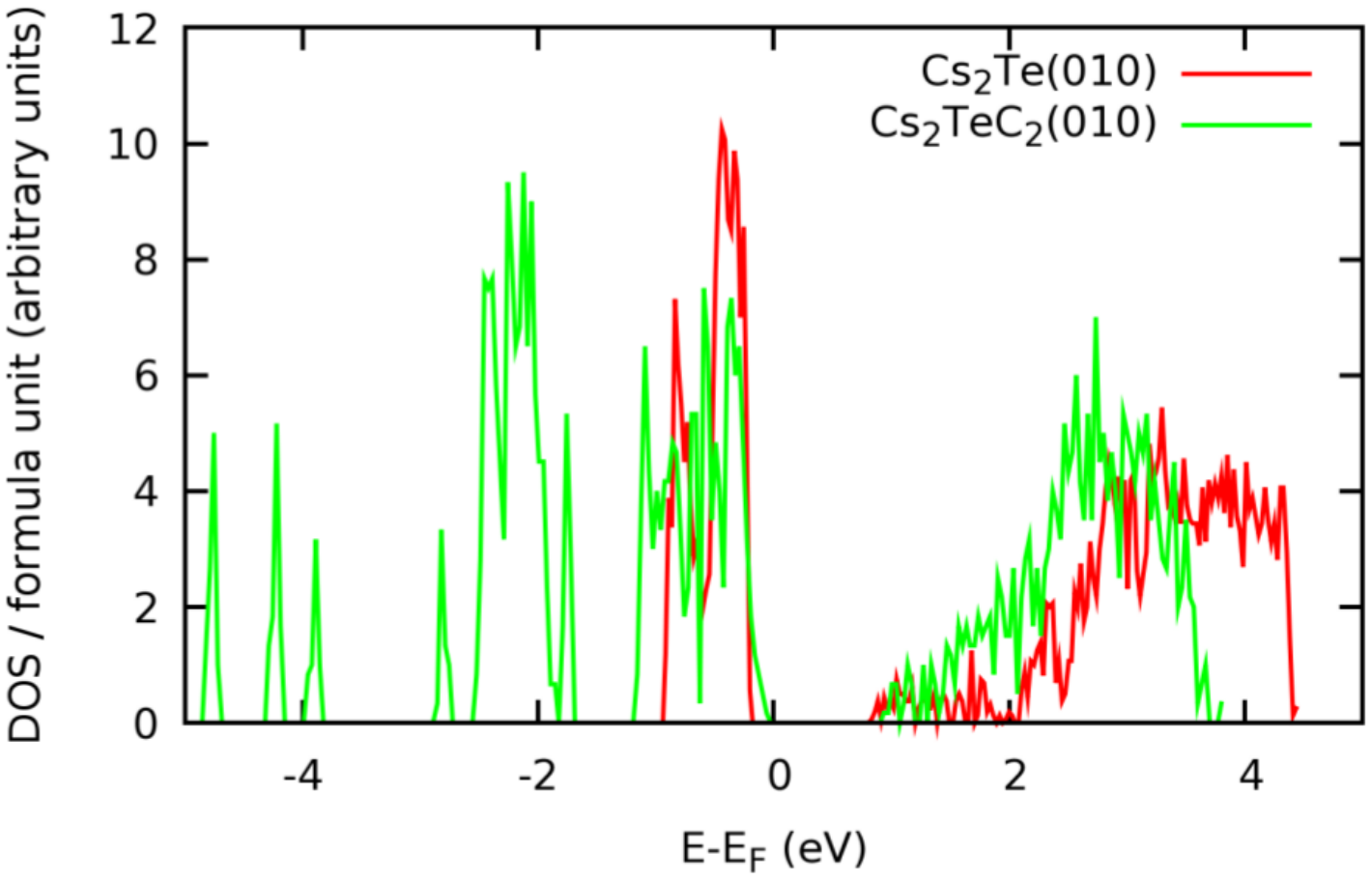
J.Z. Terdik, K. Nemeth, K. Harkay, et al, submitted for publication.

TABLE IV: Calculated (DFT) properties of photoemissive surfaces of acetylide compounds: workfunctions (Φ), bandgaps at the Γ-point $E_g(\Gamma)$ and surface energies (σ). Relaxed slabs refer to the relaxation of unrelaxed ones with the central 2 layers fixed. For h-Cs_2C_2(001) and Na_2TeC_2(010), $E_g(\Gamma) \approx 0.05$ eV has been found for a single band above E_F as well.

Compound and surface	unrelaxed Φ (eV)	unrelaxed $E_g(\Gamma)$ (eV)	unrelaxed σ (eV/Å^2)	relaxed Φ (eV)	relaxed $E_g(\Gamma)$ (eV)	relaxed σ (eV/Å^2)
o-Cs_2C_2(010)	2.80	1.25	0.023	-	-	-
h-Cs_2C_2(001)	2.56	1.14	0.027	-	-	-
Na_2PdC_2(001)	3.58	1.13	0.067	-	-	-
Na_2PdC_2(110)	3.73	1.65	0.029	4.17	2.34	0.024
Na_2PdC_2(010)	2.65	1.91	0.019	2.68	2.45	0.017
Cs_2PdC_2(001)	2.90	1.43	0.046	-	-	-
Cs_2PdC_2(110)	2.73	0.88	0.026	2.73	1.16	0.022
Cs_2PdC_2(010)	1.33	0.78	0.015	2.03	1.74	0.013
Na_2TeC_2(001)	3.40	1.03	0.029	-	-	-
Na_2TeC_2(110)	3.80	0.91	0.025	4.67	2.04	0.009
Na_2TeC_2(010)	2.75	1.43	0.015	2.68	1.34	0.015
Cs_2TeC_2(001)	3.71	1.86	0.022	-	-	-
Cs_2TeC_2(110)	2.77	0.77	0.020	2.98	1.38	0.019
Cs_2TeC_2(010)	1.71	1.00	0.013	2.44	1.63	0.009

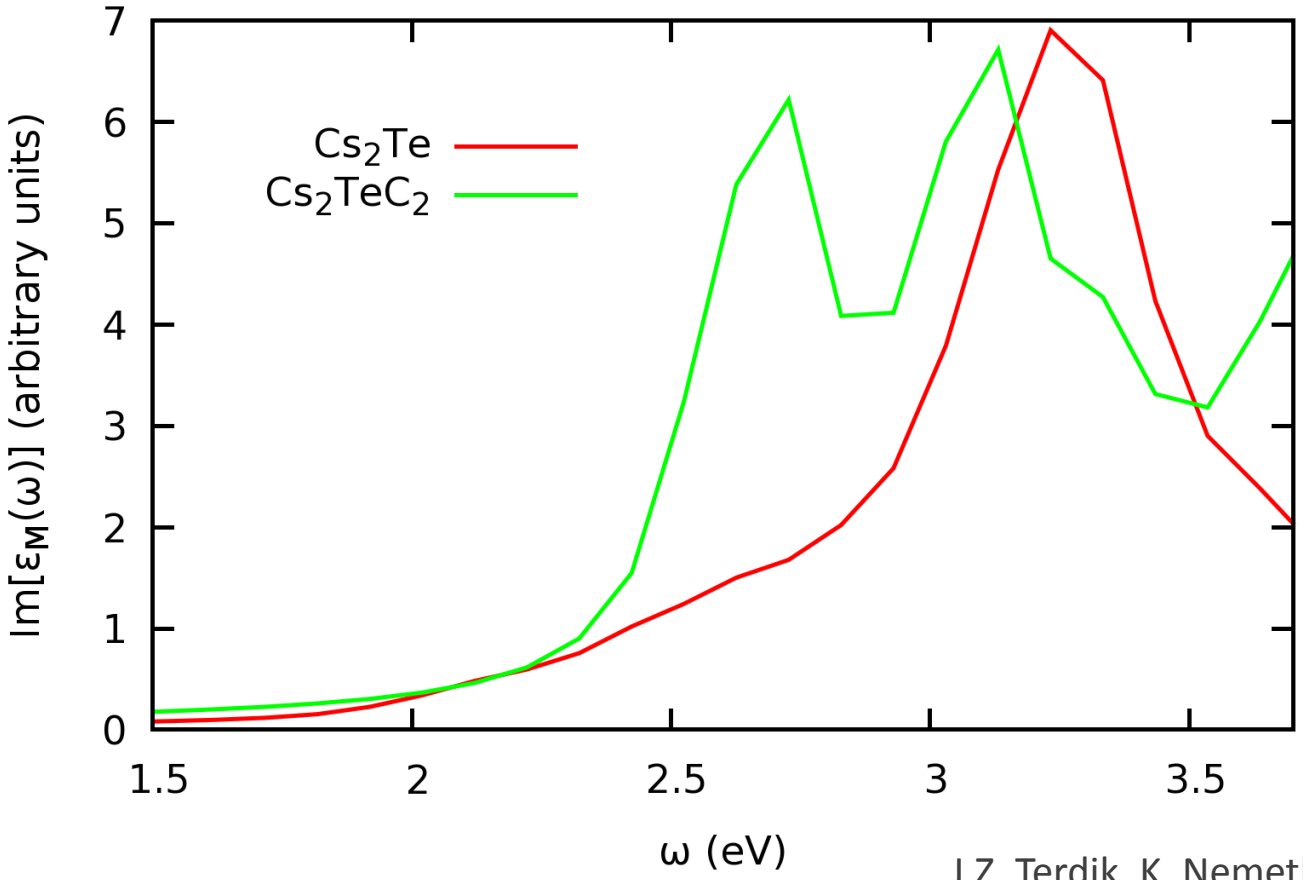
Density of states (DOS)

DOS of Cs_2TeC_2 is comparable to Cs_2Te; computed using PWSCF (Terdik, Nemeth).



QE estimation

- Computed the optical absorption spectra (i.e., imaginary part of dielectric constant) using YAMBO (Nemeth).
- QE of Cs_2TeC_2 expected to be comparable to Cs_2Te.



J.Z. Terdik, K. Nemeth, K. Harkay, et al, submitted for publication.

Synthesis/Characterization

- Collaborations in place, but we welcome more!
  - PPNL working on MgO-Ag system (see W. Hess's talk)
  - IIT-ANL working on ternary acetylide systems. *Patent application in process, K. Nemeth et al*.
- Two modes are possible for synthesis/characterization:

	pros	**cons**
separated	Specialized expertise: consistent cathode quality/performance.	Load-lock sample transport needed.
combined in one system	No transport needed.	Long learning curve to achieve consistent quality/perform.

- Preference for option 1 (ternary acetylides):
  - Local Cs_2Te expert: Z. Yusof and colleagues (ANL/AWA) (QE 10%).
  - Expertise in synthesis and surface science and instrumentation is distributed (ANL/APS, ANL/ MSD, IIT, LBNL, etc.)

Characterization

Goal:

- Was desired compound synthesized? X-ray diffraction, XPS, …
- Does it perform as predicted? Workfunction, QE, ARPES (intrinsic emittance), …

Tools available or planned:

- Load lock system (vacuum hardware, sample-transfer hardware)
- Kelvin probe (workfunction)
- ARPES at LBNL/ALS beamline (being commissioned, G. Lebedev)
- ARPES at ANL, IIT (under development)

Formation of Na_2PdC_2 as observed by X-ray diffraction in the traditional synthesis. [U. Ruschewitz, Z. Anorg. Allg. Chem. 632,705 (2006)].

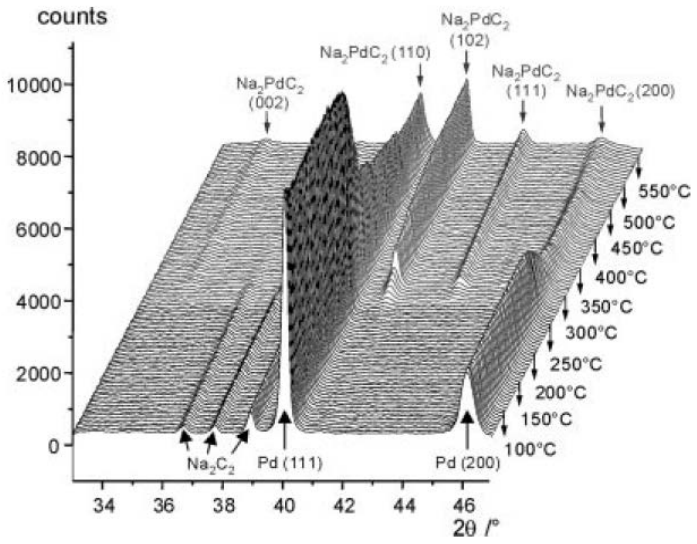
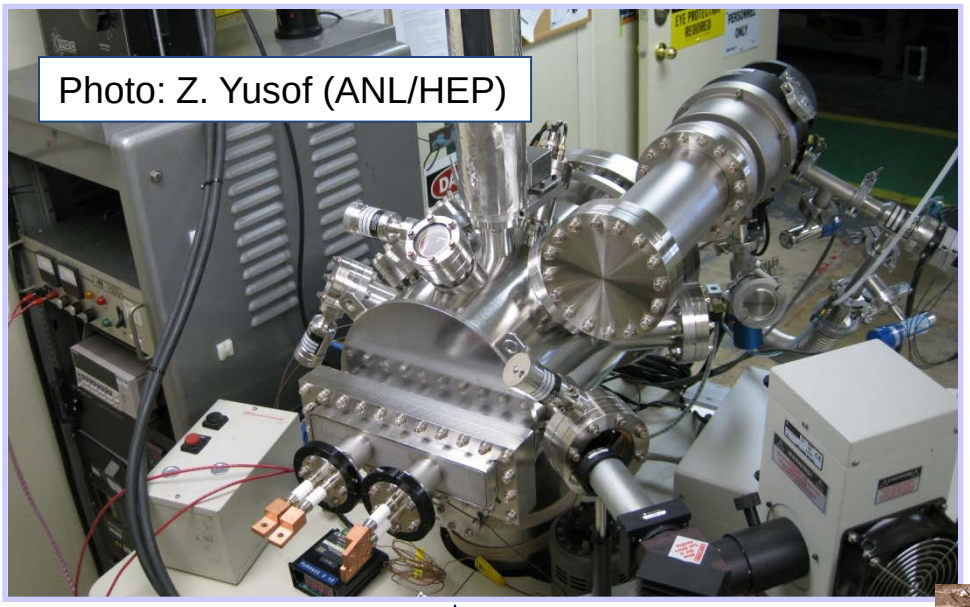


Fig. 1 Formation of Na_2PdC_2 from Na_2C_2 and palladium with increasing temperature as measured on an X-ray powder diffractometer (Huber G644, CuKα_1 radiation). The reflections of the starting materials and the product are assigned.

Cs_2Te Photocathode – Fabrication and Vacuum Transfer



Cs_2Te photocathode deposition system

Photocathode plug/substrate (Mo)

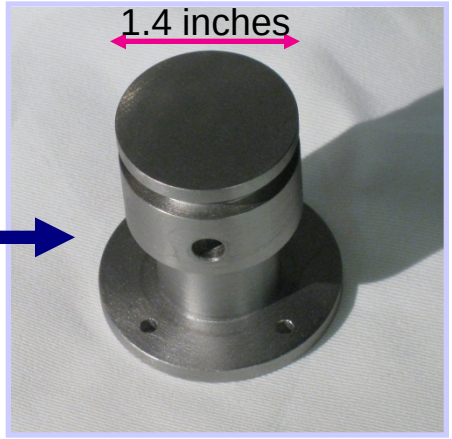
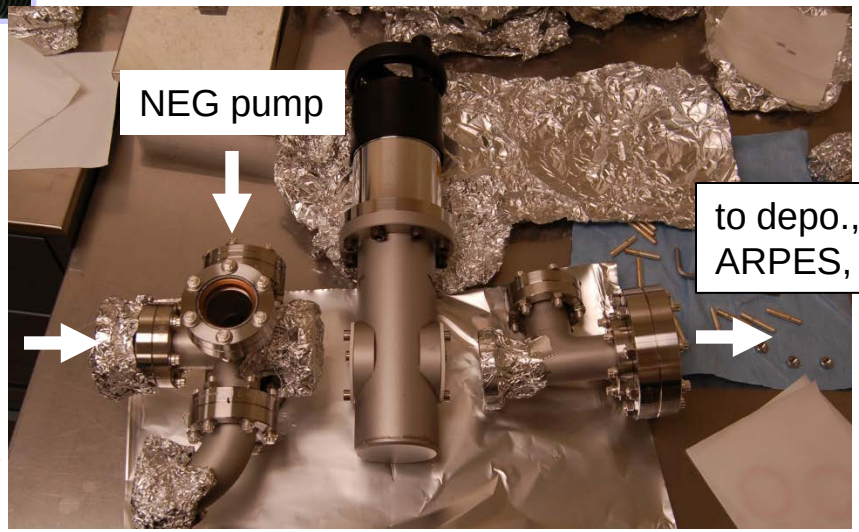


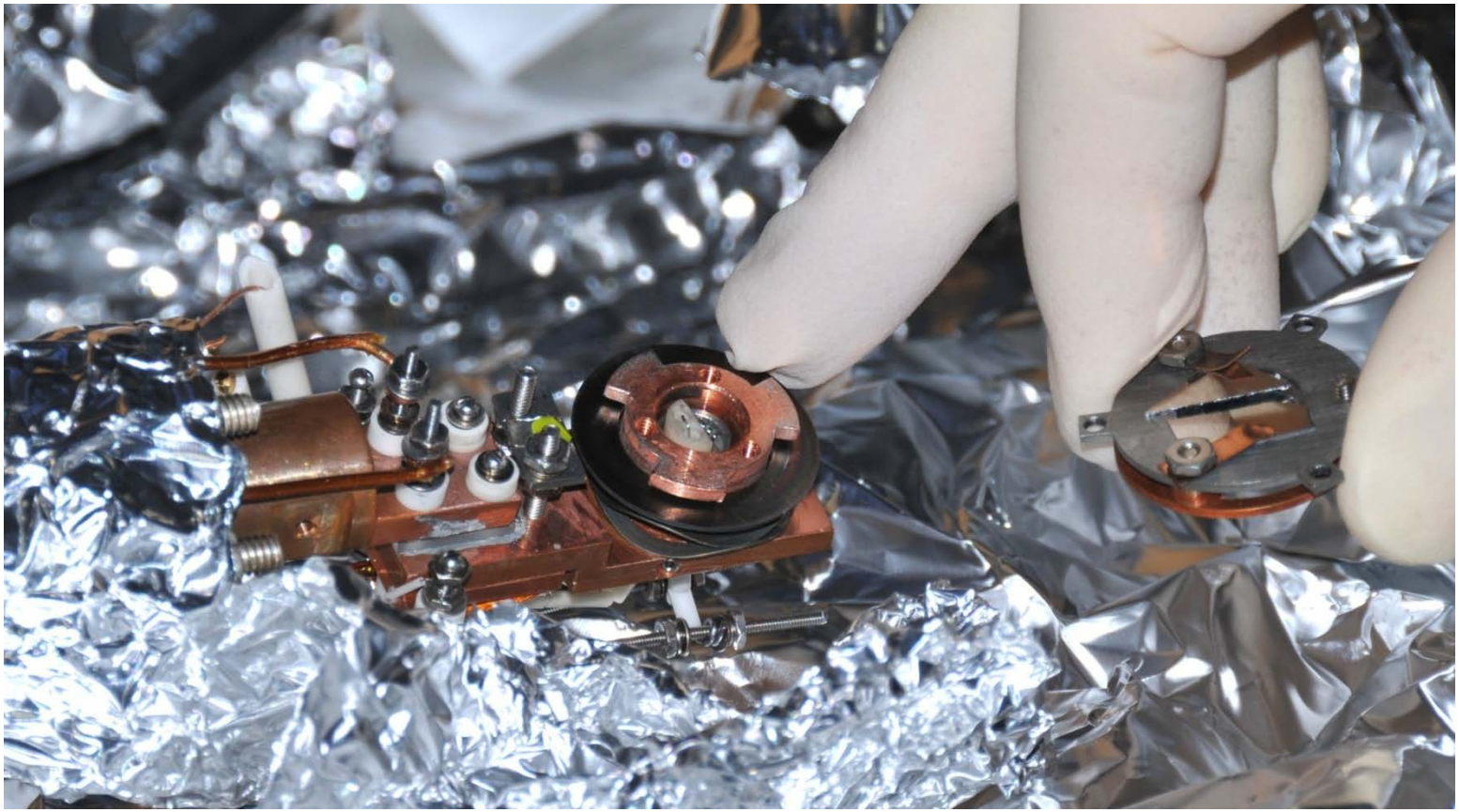
Photo: Z. Yusof



Portable UHV vacuum load-lock system

Photo: D. Velazquez (IIT)

Cross-compatible sample transfer hardware to be installed inside load-lock



Example: designed by R. Rosenberg.

FY12 Plans

1: Test transfer & characterization of Cs_2Te

- Transfer Cs_2Te from ANL to LBNL
- ALS sample transfer hardware to be adapted to AWA Cs_2Te chamber and load-lock system.
- Measure intrinsic emittance in ARPES TOF system being installed on ALS (G. Lebedev).

2: Ternary acetylide

- Synthesize Cs_2TeC_2 or other ternary acetylide at IIT and characterize at APS (QE, workfunction, structure).
- Synthesize/transfer Cs_2TeC_2 and characterize in LBNL and ANL or IIT ARPES systems.

Beyond FY12:

- Transfer to and characterize performance in an electron gun.
- Where? LBNL APEX VHF injector, Jlab, APS Injector Test Stand, …

Resources

2009 Sept, DOE/BES Workshop on Accelerator Physics of Future Light Sources

D. Dowell et al., Nucl. Instrum. Meth A 622, 685 (2010).

Physics of Photocathodes for Photoinjectors

2010, Oct 12-14, BNL

http://www.bnl.gov/pppworkshop/

2012, Oct 8-10, Cornell

Announcement & web site posted soon

Several European photocathode workshops

Summary

- Ultra-bright photocathodes are a key technology for future light sources.
- We are thinking "outside the box" to tune photocathode properties to enhance future light source performance. More work is needed.
- MgO-Ag system has been shown experimentally to reduce the workfunction; calculations predict ultra-low transverse emittance.
- "Designer" acetylated Cs_2Te – Cs_2TeC_2 – and other systems in this class are predicted to have low workfunctions in the visible to near-IR range. The QE of Cs_2TeC_2 is predicted to be comparable to Cs_2Te. The rod-perpendicular orientation may exhibit low emittance.
- Synthesis of both ideas is ongoing through collaborations (PNNL and IIT/ANL, respectively) and additional collaborators are welcome.
- Characterization using surface science techniques is planned at APS and at LBNL/ALS beamline ARPES system; load-lock transfer needed.
- Characterization in a gun of promising cathodes is desired; load-lock transfer needed.