UNIVERSITÄT GREIFSWALD
Wissen lockt. Seit 1456

Electron emission and backscattering from surfaces+

Franz Xaver Bronold

Institute of Physics

University Greifswald

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Outline

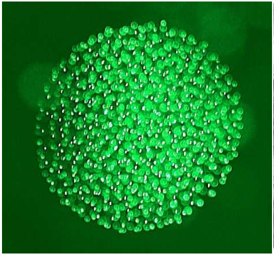
1. **Motivation**
2. **Calculational approach**
3. **Numerical results**
4. **Conclusion**

Motivation

Secondary electron emission – Why should we care?

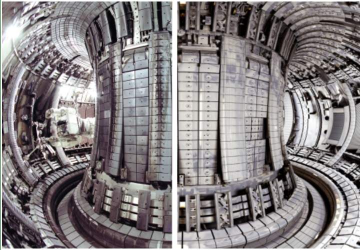
man-made plasmas are bounded by confining solid walls

dusty plasmas



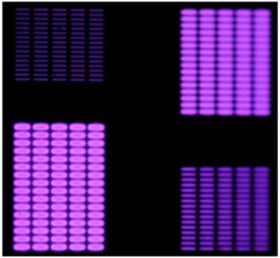
A. Melzer

fusion plasmas



Schorn, FZ Juelich

microdischarges



Kulsreshath, PSST (2014)

- plasma interacts with solids
- ion-, radical-, and **electron-solid interaction** $\rightarrow \alpha, Y_e^*,$ **(**S_e**,** R_e**,** $Y_e \rightarrow S, R, Y$**)**
- electron impact energies depend on plasma context
- focus of presentation: **very low electron impact energies (**$< 50\,\mathrm{eV}$**)**

Solid-state-based microdischarges

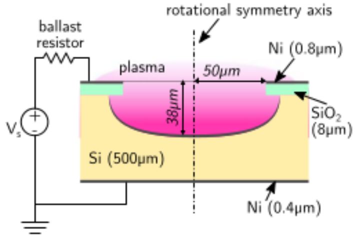
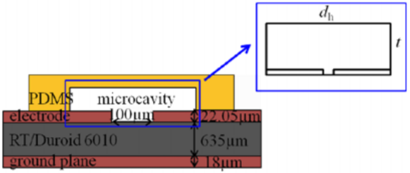


Figure 1. Schematic of a single silicon based MHCD reactor produced by MEMS manufacturing technology.

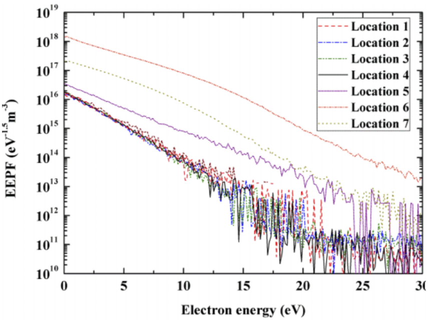
S. Iseni *et al.*, PSST. **28**, 065003 (2019)

- V_s typically a few 100 eV
- plenty of e^- interacting with solid boundaries at $E < 50\,\mathrm{eV}$
- need R_e, S_e, and Y_e for **very low impact energies** and variety of materials



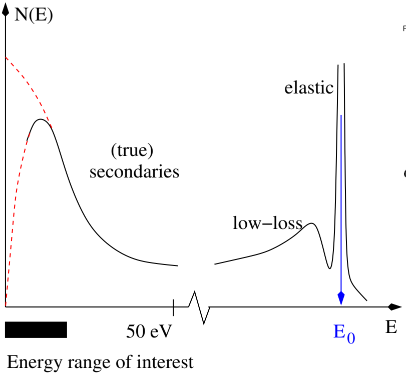
J. Tang *et al.*, AIP Adv. **6**, 045016 (2016)

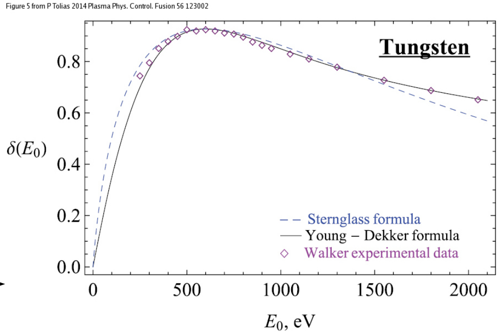
location 1–4 along electrode



See also, J. Choi *et al.*, IEEE Trans. Plasma Science **35**, 1274 (2007)

Very low electron impact energies – challenge



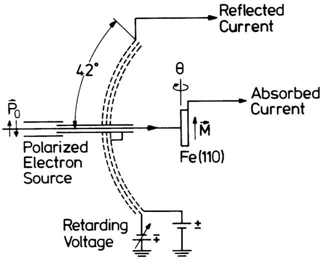


From P. Tolias, PPCF **56**, 123002 (2014).

- $E_0 < 50\,\mathrm{eV} \Rightarrow$ true secondaries and inelastically scattered primaries **not clearly separable**
- **solid state effects matter** (surface potential, electronic structure)
- established empirical formulae for $\delta(E_0)$ [$\rightarrow Y(E_0)$] not applicable

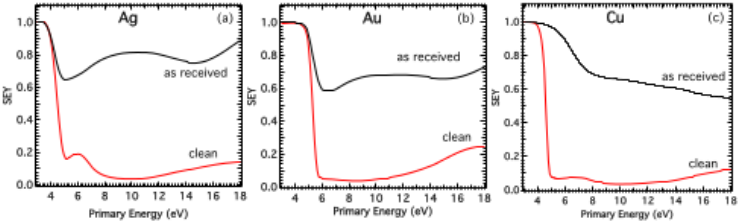
$\Rightarrow Y$, R, and S have to be calculated and measured

Measuring secondary electron emission yields – beam experiments



- sophisticated electron spectroscopy and vacuum equipment
- angle and energy resolved measurements
- control over surface quality (single crystals)
- but: not applicable to plasma environment

M. S. Hammond *et al.*, PRB **45**, 6131 (1992)



Focus on "clean" (well characterized) samples! L. A. Gonzalez *et al.*, AIP Advances **7**, 115203 (2017)

Measuring electron sticking coefficients – electric probe experiments

V. I. Demidov, S. F. Adams, I. D. Kaganovich, M. E. Koepke, I. P. Kurlyandskaya, Phys. Plasma **22**, 104501 (2015)

1 & 2=clean & dirty Mo; 3=dirty Mo (Bronshtein), 4=Cu (Cimino)

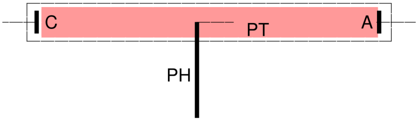
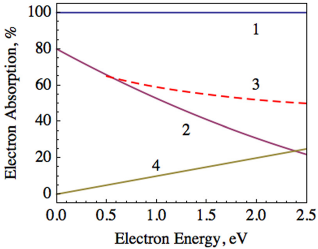


FIG. 3. Schematic of the experimental device. A glass tube has a diameter of 35 mm and length of 30 cm with cold cathode (C) and anode (A). A probe tip (PT) is situated along the tube axis, and probe holder (PH) is perpendicular to the tube axis.



- Ne afterglow plasma, **heavy particle collisions generate e^- beam with E_k**

$$\mathrm{Ne}^* + e^- \to \mathrm{Ne} + e^-(E_k)$$

- probe current modified by electron reflection $I_d(V) = I_c(V)G(eV)$

$$G(U) = 2\int_0^{\pi/2} d\theta \sin\theta\cos\theta\, S_e(E-U,\theta)$$

- possibility to extract $S_e = 1 - Y_e$
- interesting operando method, but how well characterized are surfaces?

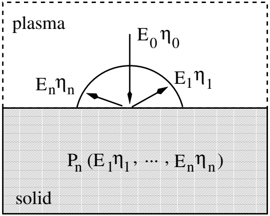
Calculational approach

Before we get started

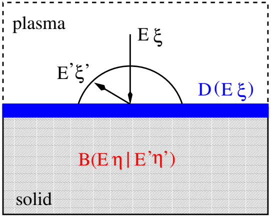
Connection to plasma modeling

→ electron-solid interaction encapsulated in BC
→ two fundamentally different approaches to do that

probabilistic emulation [1]



our (physical) approach



- **input:** $Y_e = Y_{\rm el} + Y_{\rm inelastic} + Y_{\rm ts}$, dY_e/dE
- **output:** $P_n \Leftrightarrow$ **prob. for event in particle-based simulation**

- **input:** microphysics of interface (elementary excitations, $E(\vec{k})$, ...)
- **output:** Y_e, $dY_e/dE \Rightarrow$ **surface scattering kernel=BC for e^- BE** $f_e^> = R_e \circ f_e^<$

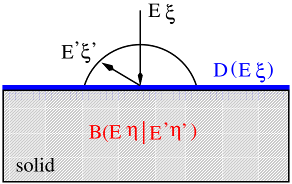
[1] M. A. Furman and M. T. F. Pivi, Phys. Rev. Spec. Topics - Accelerator and Beams, **5**, 124404 (2002)

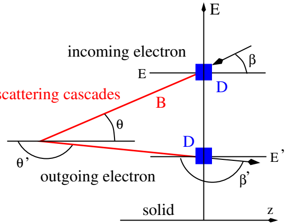
Stages of development of our (physical) approach

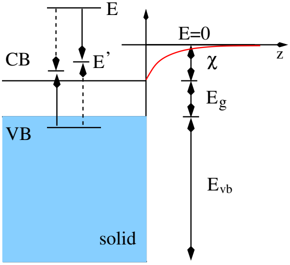
- initially, we applied our approach to dielectrics with $\chi > 0$ [1]
  $\rightarrow$ conditional probability construction to obtain B from Q
  $\rightarrow$ step potential, scattering on optical phonons and surface defects
  $\rightarrow$ $E < E_g \Rightarrow$ no impact ionization
  $\rightarrow$ linearized embedding equation for $Q \Rightarrow$ algebraic recursion relations
- then, we generalized our approach substantially and applied it to metals [2]
  $\rightarrow$ still conditional probability construction for B
  $\rightarrow$ image step with Bragg scattering, energy gaps at surface
  $\rightarrow$ ei, ep, ee, and scattering on surface defects
  $\rightarrow$ no linearization of embedding equation but quasi-isotropic approximation
  $\Rightarrow$ set of linear integral equations instead of algebraic equations
- **now, we applied it to semiconductors with $\chi > 0$ [3]**
  $\rightarrow$ conditional probability construction for B abandoned
  $\rightarrow$ image step, ei, ep, and impact ionization (hence, $E > E_g$ now allowed)
  $\rightarrow$ no linearization and no quasi-isotropic approximation
  $\Rightarrow$ full embedding equation for $Q \Rightarrow$ Ricatti and a set of Sylvester equations

[1] FXB and H. Fehske, PRL **115**, 225001 (2015); ibid. PPCF **59**, 014011 (2017); [2] ibid. JAP **131**, 113302 (2022); [3] FXB and F. Willert, arXiv:2309.00534 and submitted (2023).

Calculational approach – cartoon







Notation: scattering from $(E, \eta) \rightarrow (E', \eta')$ model for semiconductor

So far: chemically clean surfaces;
surface chemistry due to plasma could be buried in **D**

Emission yield for structurally perfect surface

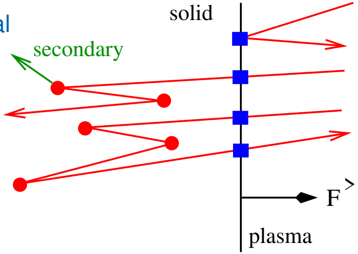
$$Y(E,\xi) = \int_0^E dE' \int_0^1 d\xi' R(E,\xi|E',\xi')$$

$$\underbrace{R(E,\xi|E',\xi')}_{\text{electron surface scattering kernel}} = R(E,\xi)\delta(E-E')\delta(\xi-\xi') + \Delta R(E,\xi|E',\xi')$$

Calculational approach – electron surface scattering kernel

Electron-solid interaction

- qm transmission through interface potential
  electron mass mismatch
  $V(z)$ across interface
  energy gaps
- scattering cascades inside solid
  elastic $\&$ inelastic processes
  electron multiplication
  band structure
- **electron surface scattering kernel** $R \Rightarrow$ emission yield Y $\&$ BC for BE



$$F^>(E',\xi') = \int_0^\infty dE \int_0^1 d\xi F^<(E,\xi) R(E,\xi|E',\xi')$$

$$R(E,\xi|E',\xi') = R(E,\xi)\delta(E-E')\delta(\xi-\xi') + \Delta R(E,\xi|E',\xi')$$

$$\Delta R(E,\xi|E',\xi') = \frac{E'}{E'+\chi}\frac{\xi'}{\eta'}\rho(E')\Theta(E-E')D(E,\xi)B(E,\eta(\xi)|E',\eta(\xi'))D(E',\xi')$$

Calculational approach – alternative expression for Y [1]

Emission yield $Y(E,\xi)$ for an electron hitting perfect ($C=0$) or irregular ($C\neq 0$) surface with (E,ξ)

- **irregular** surface $\rightarrow \int_0^1 d\xi' (...)$
- **qm transmission** through surface potential $\rightarrow \mathbf{D(E,\xi) = 1 - R(E,\xi)}$
- **internal scattering cascade** $\rightarrow B(E\eta|E'\eta') \rightarrow$ invariant embedding principle

$$Y(E,\xi) = 1 - S(E,\xi)$$

$$S(E,\xi) = \frac{D(E,\xi)}{1+C/\xi}\left[1-\mathcal{E}(E,\xi)\right] + \frac{C/\xi}{1+C/\xi}\int_0^1 d\xi' D(E,\xi')\left[1-\mathcal{E}(E,\xi')\right]$$

$$\mathcal{E}(E,\xi) = \int_{\eta_{\mathrm{min(E)}}}^1 d\eta' \int_{E_{\mathrm{min}}(\eta')}^E dE' \rho(E') B(E\eta(\xi)|E'\eta') \overline{D}(E',\xi(\eta'))$$

$$\overline{D}(E,\xi) = \frac{D(E,\xi)}{1+C/\xi} + \frac{C/\xi}{1+C/\xi}\int_0^1 d\xi' D(E,\xi') \;\; , \quad C \sim |M|^2 n_{\mathrm{surface\,defects}}$$

Limiting cases: $C\rightarrow 0$ (perfect) and $C\rightarrow\infty$ (very irregular)

[1] FXB and H. Fehske, PRL **115**, 225001 (2022); ibid. PPCF **59**, 014011 (2017)

Calculational approach – capabilities/options

overall

- irregular surface $\Leftrightarrow$ (elastic) surface defects

surface transmission function D

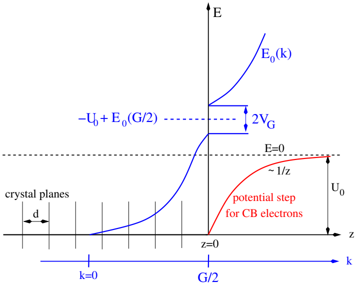
- image step plus Bragg gap (two band model)

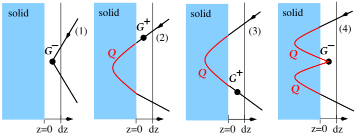
bulk transition rates $W^{\pm}$

- (statically screened) el-el scattering
  $\rightarrow$ two final states
- (quasi-elastic) electron-phonon scattering
- incoherent elastic scattering by ion cores
  $\rightarrow$ pseudo-potentials for ion cores

invariant embedding equations for $Q \Rightarrow B$

- decoupling of E and $\eta \Leftrightarrow$ quasi-isotropic approximation (QIA) [1]
- **now: numerical solution without QIA** [2]





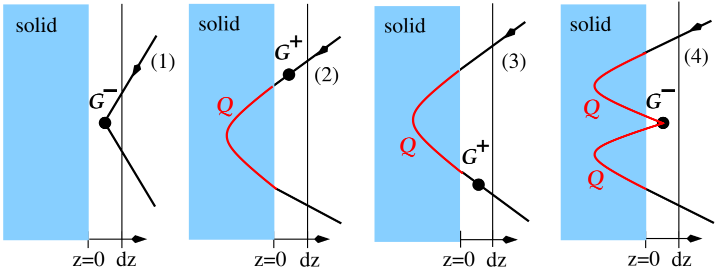
- $\hat{=}\ G^{+}$ or G^{-} with $G^{\pm} = W^{\pm}/\eta v$

[1] FXB and H. Fehske, JAP **131**, 113302 (2022); [2] FXB and F. Willert, arXiv:2309.00534 and submitted (2023).

Calculational approach – invariant embedding principle

Dashen (1964): Adding an infinitesimally thin layer of the same material to a thick surface should not alter $Q(E\eta|E'\eta')$.

R. Dashen, Phys. Rev. **134**, A1025 (1964)



$\bullet \hat{=} G^+$ or G^- with $G^\pm = W^\pm/\eta v$

$$dQ = \Big\{ G^- + G^+ \circ Q + Q \circ G^+ + Q \circ G^- \circ Q - \Big(\frac{\Pi}{\eta} + \frac{\Pi'}{\eta'}\Big) Q \Big\} dz = 0$$

$$(A \circ B)(E\eta|E'\eta') = \int_{E'}^{E} dE'' \int_0^1 d\eta'' \rho(E'') A(E\eta|E''\eta'') B(E''\eta''|E'\eta')$$

Calculational approach – kernels

Scattering rates and the like – Golden Rule $(E,\eta) \rightarrow (E',\eta')$

$$G^{\pm}(E\eta|E'\eta') = \frac{W^{\pm}(E\eta|E'\eta')}{v(E)\eta} \qquad \Pi(E) = \frac{\Gamma(E)}{v(E)}$$

$$W^{\pm}(E\eta|E'\eta') = \int_0^{2\pi} d\phi\, W(E\eta|E'\eta';\phi)$$

$$\Gamma(E) = \int_{-\chi}^{E} dE'' \int_0^1 d\eta'' \rho(E'')[W^+(E\eta|E''\eta'') + W^-(E\eta|E''\eta'')]$$

$$W^{\pm}(E\eta|E'\eta') = W^{\pm}_{\mathrm{el}}(E\eta|E'\eta') + W^{\pm}_{\mathrm{ee}}(E\eta|E'\eta')$$

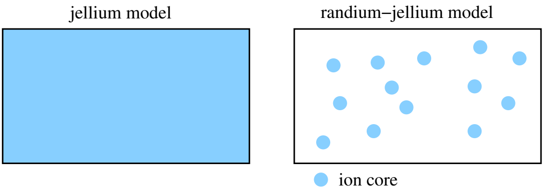
Embedding equation for $Q(E\eta|E'\eta')$ **to be solved**

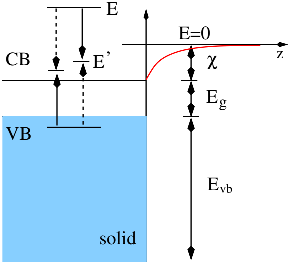
$$S \circ Q + Q \circ S = G^- + G^+ \circ Q + Q \circ G^+ + Q \circ G^- \circ Q$$

$$S(E,\eta|E',\eta') = \frac{\Pi(E)}{\eta}\delta(E-E')\delta(\eta-\eta')$$

Calculational approach – randium jellium model for ion cores [1]

- incoherent scattering on randomly distributed ion cores
- quasi-elastic electron-phonon scattering
- impact ionization across bulk energy gap





$$W^{\pm}_{\rm elastic}(E,\eta|E',\eta') = W^{\pm}_{\rm ep}(E,\eta|E',\eta') + W^{\pm}_{\rm eic}(E,\eta|E',\eta')$$

$$W^{\pm}_{\rm ep}(E,\eta|E',\eta') = \frac{M^2}{(2\pi)^2}[1+2n_B(\omega_{\rm LO})]\delta(E-E')\Theta(E_{\rm th}-E)$$

$$W^{\pm}_{\rm eic}(E,\eta|E',\eta') = \frac{1}{(2\pi)^2 n_{\rm ion}}\langle|U_{\rm ps}(g^{\pm})|^2\rangle_\Phi \delta(E-E')\Theta(E-E_{\rm th})$$

$$W^{\pm}_{\rm impact}(E,\eta|E',\eta') = \mathcal{W}(E,T,1|E',T',\pm1) \quad \text{Monte Carlo integration required}$$

[1] For details see, FXB and F. Willert, arXiv:2309.00534 and submitted (2023).

Calculational approach – numerics

Adopting Shimizu and Mizuta's approach of solving embedding-type equations in nuclear transport theory. A. Shimizu and H. Mizuta, J. Nucl. Sci. Technol. **3**, 57 (1966)

(i) Discretization of energy space

$$A_{nm}(\eta|\eta') = \int_n dE \int_m dE' A(E,\eta|E',\eta') f_n(E) \quad , \quad f_n(E) = 1/\Delta E_n$$

$\Rightarrow$ Ricatti/Sylvester equations for $Q_{nm}(\eta|\eta')$

$$S_n * Q_{nn} + Q_{nn} * S_n = G^-_{nn} + Q_{nn} * G^-_{nn} * Q_{nn} + G^+_{nn} * Q_{nn} + Q_{nn} * G^+_{nn}$$

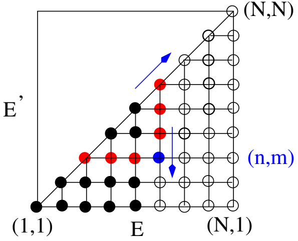
$$S_n * Q_{nm} + Q_{nm} * S_m = K^-_{nm}$$

(ii) Realizing Volterra-type structure of energy integrals

$\Rightarrow$ sampling energy space such that $Q_{kr}(\eta|\eta')$ inside K^-_{nm} known from previous step

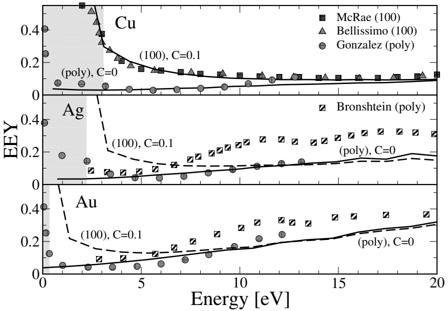
$$K^-_{nm} = \ldots + C^-_{nm} + \ldots$$

$$C^-_{nm} = \sum_{l=m+1}^{n-1} \sum_{p=m}^{l} Q_{nl} * G^-_{lp} * Q_{pm}$$



Numerical results

Results – metals (Cu, Ag, Au)



- quasi-isotropic approximation (not discussed in this talk)

FXB and H. Fehske, JAP **131**, 113302 (2022)

- **cond. prob. construction**

$$B = B(E\eta|E'\eta') = \frac{(2Q_{\rm ee} + Q_{\rm ei})(E\eta|E'\eta')}{\int_{-\chi}^{E} d\bar{E} \int_0^1 d\bar{\eta} \rho Q(E\eta|\bar{E}\bar{\eta})}$$

- successful also for W and Al

- **for Cu (100) nearly perfect agreement** between theory and experiment

E. G. McRae and C. W. Caldwell, Surf. Science **57**, 77 (1976)

A. Bellissimo, PhD thesis, Universitá degli Studi Roma Tre (2019), https://doi.org/10.5281/zenodo.3924096

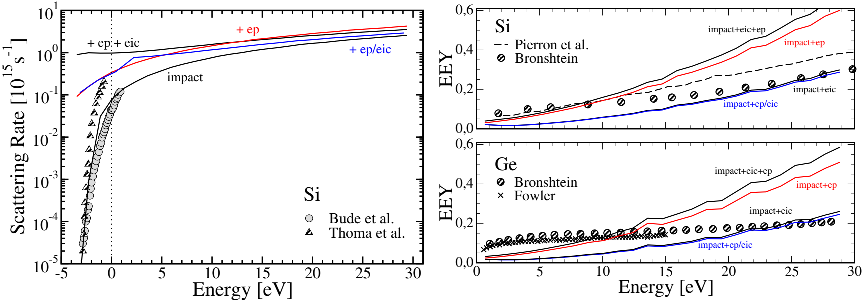
- for $E > 2\,\rm eV$ Gonzalez data for Cu, Ag, and Au [2] **fairly well reproduced**

L. A. Gonzalez *et al.*, AIP Advances **7**, 115203 (2017)

Despite quantitative success, cond. prob. construction problematic.

Wrong limit for vanishing interface potential; Flaw of construction perhaps compensated by QIA?; Under investigation.

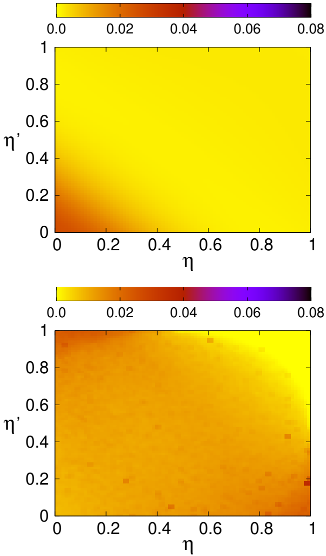
Results – semiconductors (Si, Ge) – $Y(E, \xi = 1)$



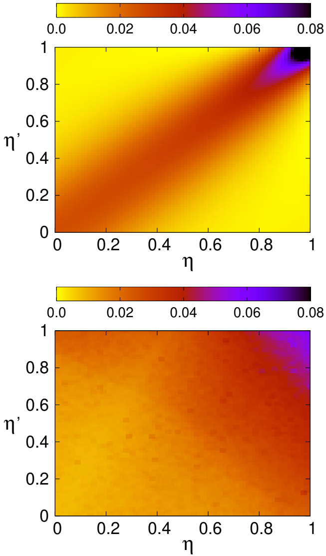
- **no cond. probability construction**: $B(E\eta|E'\eta') = Q(E\eta|E'\eta')$
- **semi-empirical description of solid** (band structure, scattering, screening)
  $\rightarrow$ parabolic VB and CB with $m^*_{\mathrm{CV}} = m^*_{\mathrm{VB}} = m_e$
  $\rightarrow$ adhoc ep/eic when $\lambda_{\mathrm{dB}} \sim a_{\mathrm{L}}$, that is, at $E_{\mathrm{th}} = -\chi + (2\pi/a_{\mathrm{L}})^2 \sim 0$ eV
  $E < E_{\mathrm{th}}$: impact+ep; $E > E_{\mathrm{th}}$: impact+eic
  $\rightarrow$ screening of ee, eic, and ep (bond vs. atomic like VB charge)
- experimental data reasonably well reproduced; **need more measurements!**
  $\rightarrow$ no tunable parameters!

Results – semiconductors (Si) – $W^{\pm}(E\eta|E'\eta')$ – angle dependence

$W^- : E = 28.8\,\mathrm{eV}, E' = 28.8, 1.8\,\mathrm{eV}$



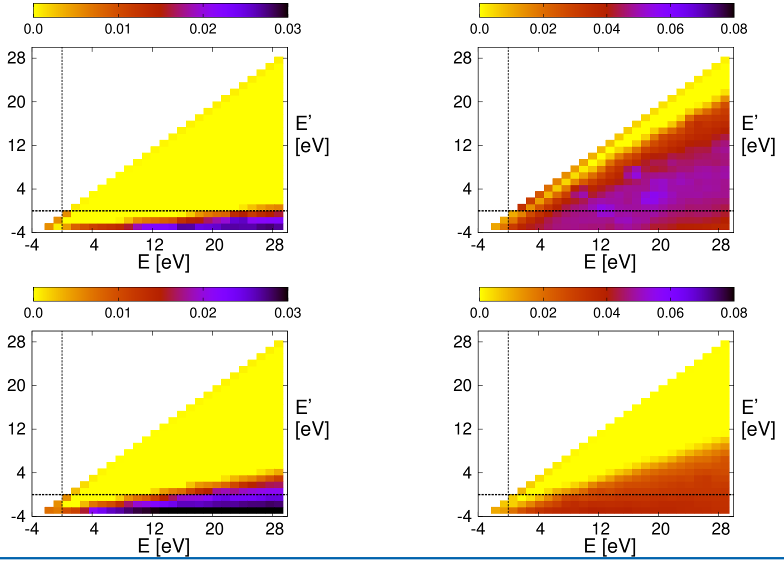
$W^+ : E = 28.8\,\mathrm{eV}, E' = 28.8, 1.8\,\mathrm{eV}$



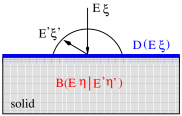
Results – semiconductors (Si) – $W^{\pm}(E\eta|E'\eta')$ – energy dependence

$W^{-} : \eta = 1, \eta' = 0.75, 0.24$

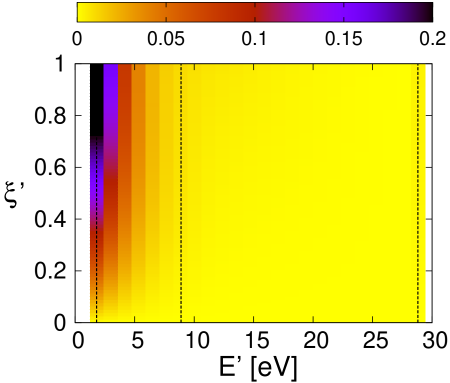
$W^{+} : \eta = 1, \eta' = 0.75, 0.24$

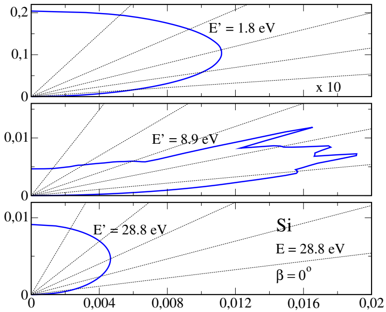


Results – semiconductors (Si) – $R(E,\xi|E',\xi')$



- primary e^-: $\xi = 1$ and $E = 28.8\,\mathrm{eV}$



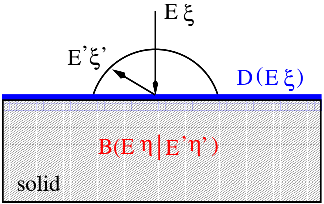


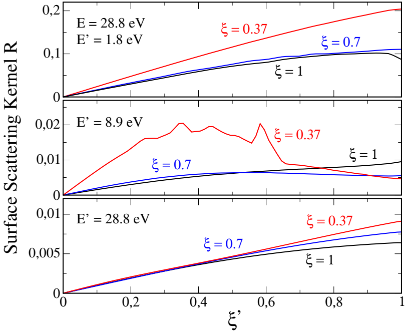
- $R(E,\xi|E',\xi')$ can be obtained for any E and ξ
  $\Rightarrow$ BC for BE which takes solid's electron microphysics into account

$$F^{>}(E',\xi') = \int_0^\infty dE \int_0^1 d\xi F^{<}(E,\xi) R(E,\xi|E',\xi')$$

Conclusion

Take home messages





- embedding approach for $Q(E,\eta|E',\eta')$ used to calculate surface scattering kernel [1] and not only emission yield [2]

  [1] FXB and F. Willert, arXiv:2309.00534 and submitted (2023); [2] FXB and H. Fehske, JAP **131**, 113302 (2022); ibid. PPCF **59**, 014011 (2017); ibid. PRL **115**, 225001 (2015).

- embedding equation for Q solved without approximation

  $\rightarrow$ **transport problem for $Q(E\eta|E'\eta')$ under control**

  $\rightarrow$ **focus now on semi-empirical description of solid**

  $\rightarrow$ **operando diagnostics of plasma-exposed surfaces required**

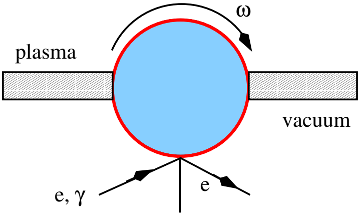
Operando surface diagnostics – How and why?

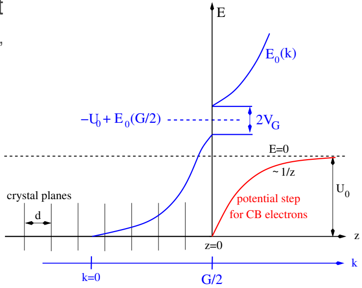
Only few experimental data for Y at $E < 20\,\mathrm{eV} \Rightarrow$ **measurements required!**

- free-standing, well-characterized surfaces – would be good
- **plasma-exposed surfaces** – better

Spinning wall setup $\rightarrow$ beam experiment

J. Guha, Y.-K. Pu and V. M. Donnelly, J. Vac. Sci. Technol. A **25**, 347 (2007)





- electronic structure of plasma-exposed surface (photoelectron spectroscopy)
  potential barriers $\rightarrow U_0$, energy gaps $\rightarrow 2V_\mathrm{G}$
- chemical and structural composition (Auger spectroscopy)
- measuring electron reflection and emission yields would be really great!