Modeling Liquid Surfaces

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- **Motivation**
- **Approach**
- **Preliminary results**

Surface-induced ordering of liquid surfaces

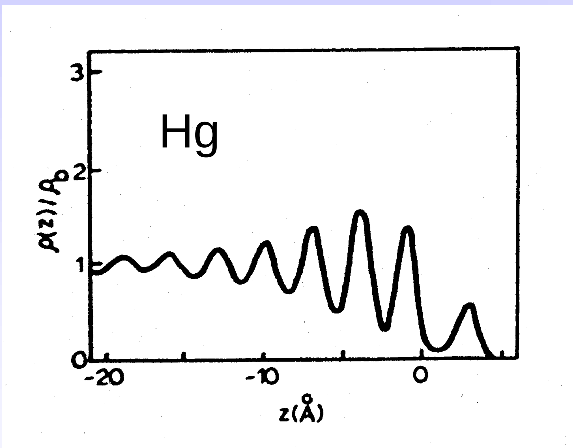
- **Predictions have been known since the late 70's and 80's**
- **Advances in synchrotron sources in last 10 years have allowed measurements to be made of the near surface**
  - **High accuracy positioning**
  - **Spectrally pure beam**
  - **In-situ oxide removal**
  - **Capillary effects accounted for**
- **Observations from many systems imply that surface ordering is a general result for liquid surfaces**
- **The effect of surface density stratification and coordination number need to be introduced into sputtering calculations for liquid metals**

Review article

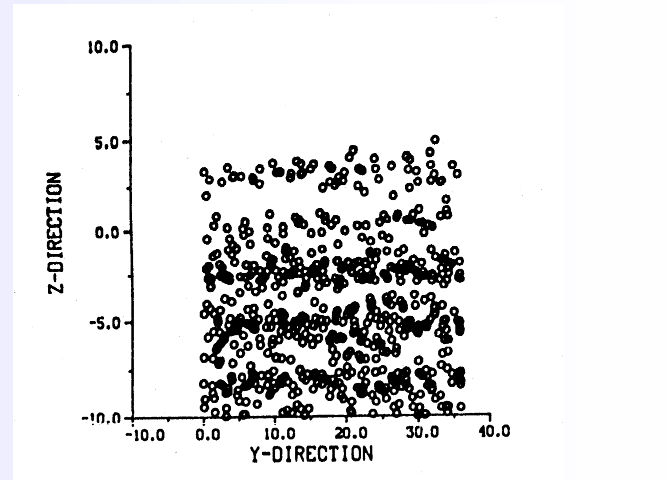
J. Penfold, Rep. Prog. Phys. 64 (2001) 777.

Early liquid surface models showed effect on self sputtering yield

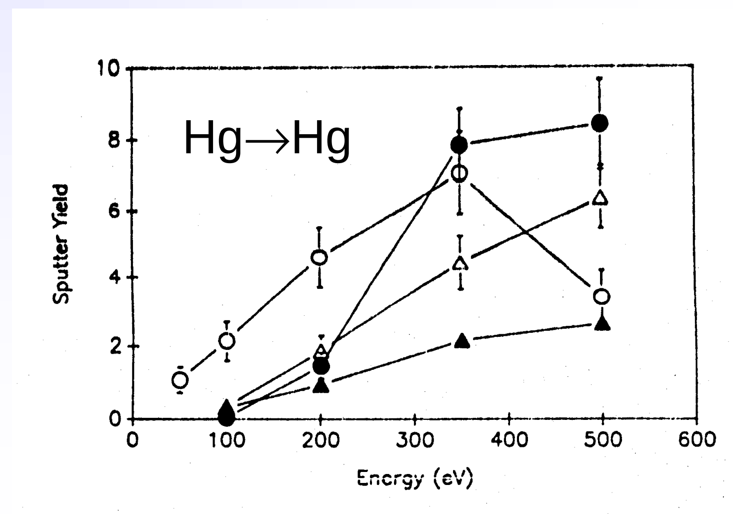
- **Stratified layer predicted by D'Evelyn and Rice (1982)**



M. P. D'Evelyn and S. A. Rice, J. Chem. Phys. 78 (1983) 5081.



- **Sputtering calculations using MD calculation and model density from above were performed by Morgan (1988)**

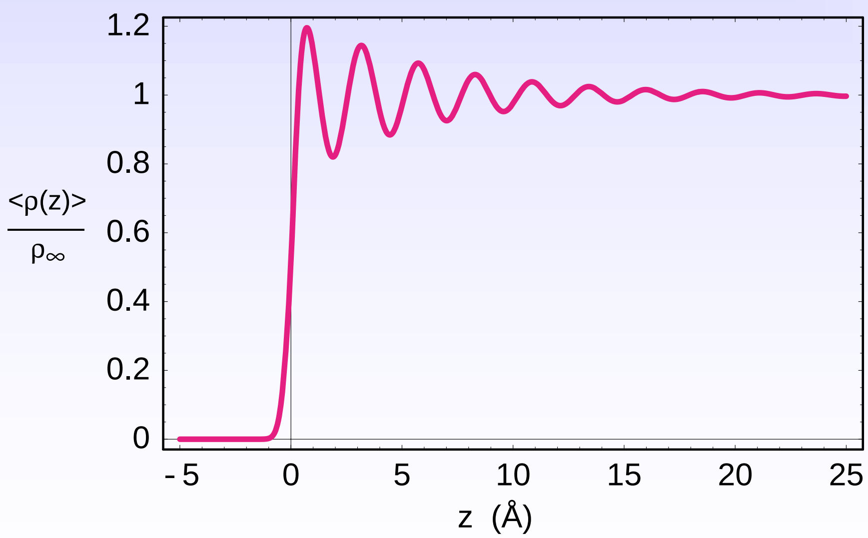


W. L. Morgan, J. Appl. Phys. 65 (1989) 1265.

Model for stratified density fit to X-ray diffraction data of liquids

- **Most recent experimental data for liquid metals is for Ga**
- **Surface density model includes layering and capillary wave broadening**
- **No lithium data or models**

$$\frac{\langle \rho(z) \rangle}{\rho(\infty)} = erf\left[\frac{z - z_0}{\sigma}\right] + \theta(z) A \sin\left(\frac{2\pi z}{d}\right) e^{-z/\zeta}$$



$$z_0 = 0$$

$$\sigma = 0.5\,\mathring{A}$$

$$A = 0.5$$

$$d = 2.56\,\mathring{A}$$

$$\zeta = 5.8\,\mathring{A}$$

M. J. Regan, et. al., Phys. Rev. Lett. 75 (1995) 2498.

Liquid sputtering model development

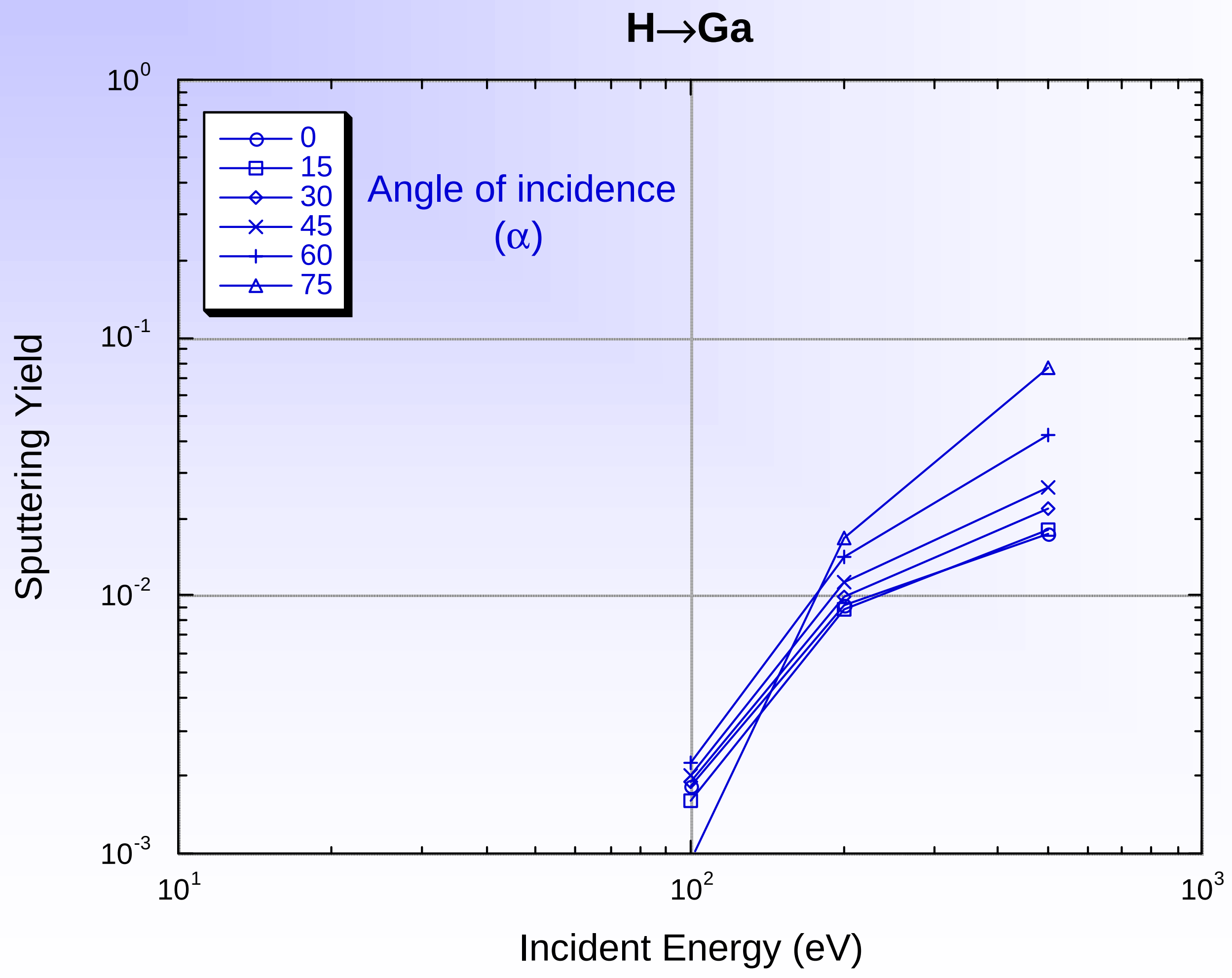
- **Use model liquid surface density function from literature**
- **Use Monte Carlo TRIM code (TRVMC98) for calculating incident and recoil trajectories and calculating collision cascade**
  - **Layer densities are stationary**
  - **Multilayer capable (present limit of 3 layers)**
  - **Compiled from source code**
  - **Vary layer density (and later surface binding energies)**
  - **Incorporate additional physics**
- **Unix Workstation (Sparc Ultra 10)**
  - **6 x 6 matrix of results requires 5 - 8 hours**

Progress to date

- **Initial calculations**
  - **Validate multi-layer code results with previous TRIM calculations (Eckstein's survey from 1993)**
  - **Initial look at effect of one period of the surface layer structure (on semi-infinite bulk density)**

QuickTime™ and a
None decompressor
are needed to see this picture.

H → Ga (solid)



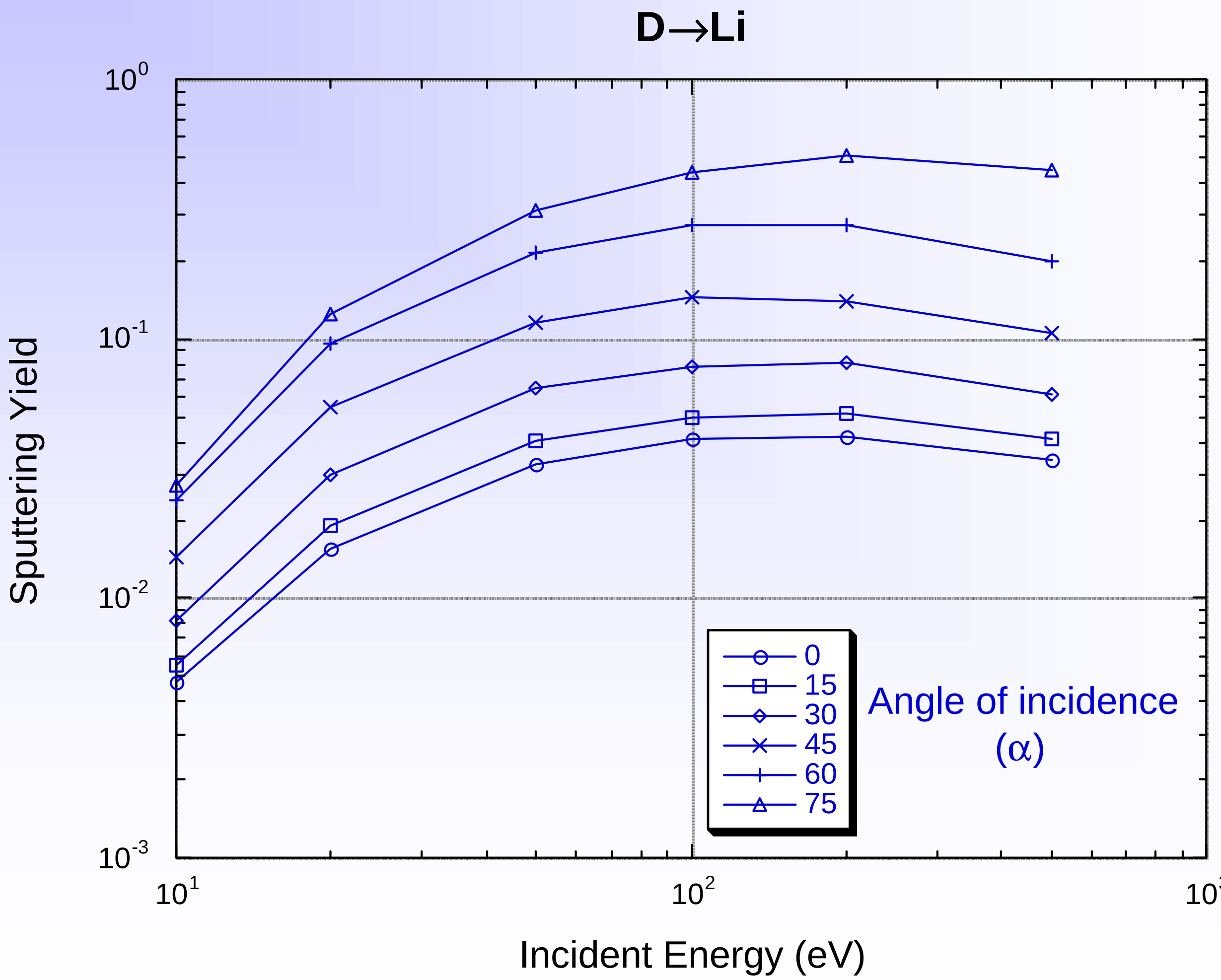
H→Ga
Sputtering Yield
Incident Energy (eV)
10^0
10^-1
10^-2
10^-3
10^1
10^2
10^3
0
15
30
45
60
75
Angle of incidence
(α)

$H \rightarrow Ga$ (liquid) $[Y_l/Y_s]$

Energy (eV)	0°	15°	30°	45°	60°	75°
10	-	-	-	-	-	-
20	-	-	-	-	-	-
50	-	-	-	-	-	-
100	0.95	1.23	0.93	1.12	0.92	1.11
200	1.00	0.92	1.05	.99	1.02	.94
500	.96	1.02	.97	1.08	1.01	1.02

Values are for 1.1/.95/1.0 profile   - = Insufficient statistics   Blue ⇒ > 15% Yield change

D → Li (solid)

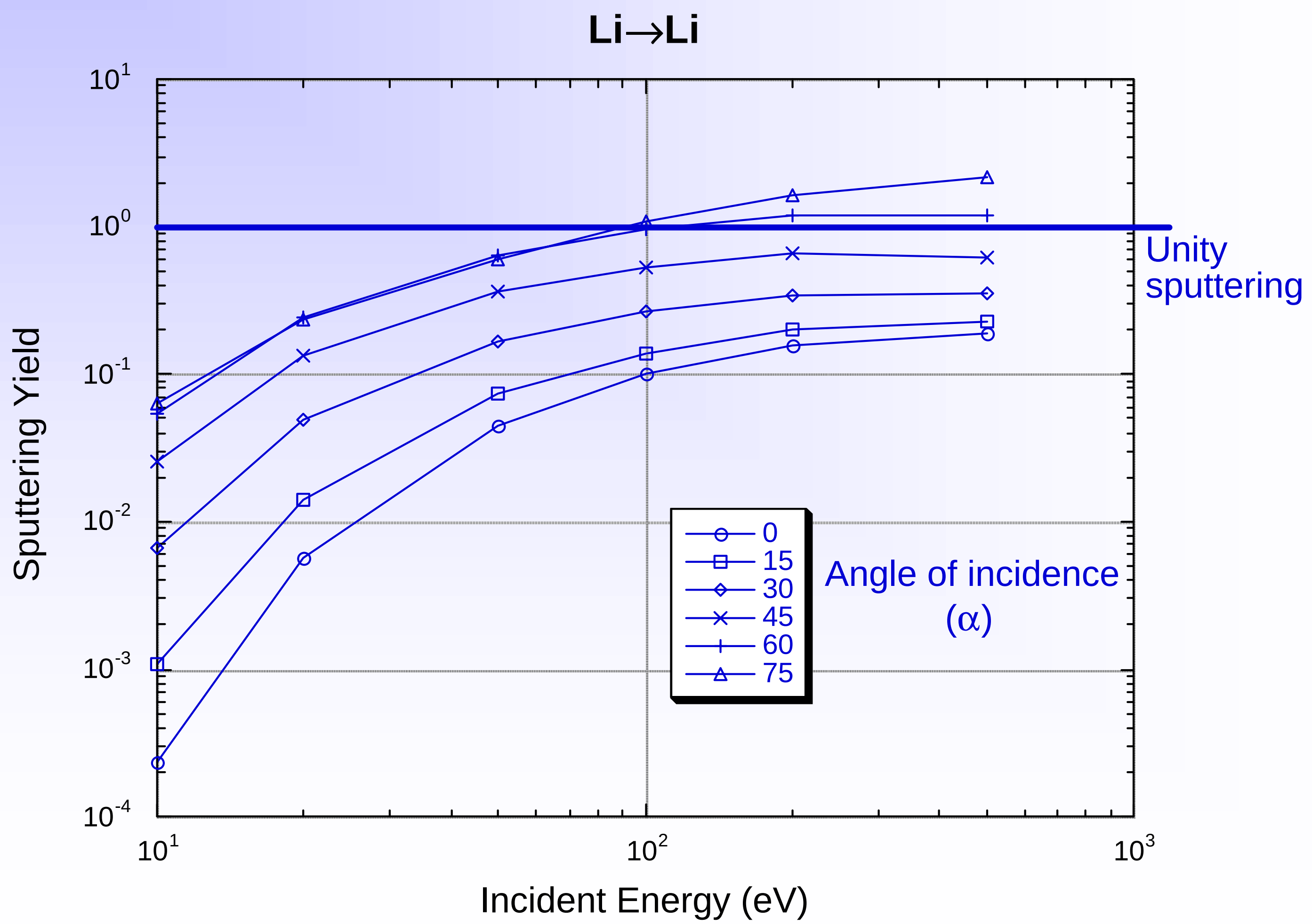

D→Li
Sputtering Yield
Incident Energy (eV)
10^0
10^-1
10^-2
10^-3
10^1
10^2
10^3
0
15
30
45
60
75
Angle of incidence
(α)

D → Li (liquid) [Y_l/Y_s]

Energy (eV)	0°	15°	30°	45°	60°	75°
10	0.92 1.16	1.01 1.02	1.15 1.17	1.01 1.06	0.99 0.96	1.03 0.93
20	1.04 1.24	1.08 1.19	1.02 1.18	1.03 1.18	1.03 1.13	1.01 0.92
50	1.05 1.22	1.02 1.15	1.02 1.17	1.04 1.18	1.04 1.15	1.01 0.97
100	1.06 1.19	1.02 1.16	1.07 1.20	1.01 1.15	1.02 1.11	1.01 1.03
200	1.04 1.21	1.02 1.16	1.01 1.12	1.04 1.14	1.01 1.09	1.02 1.05
500	1.04 1.21	1.03 1.14	0.99 1.14	1.02 1.09	1.01 1.08	1.01 1.07

Top line = 1.1/.95/1.0 profile Bottom line = 1.5/.75/1.0 profile Blue ⇒ > 15% Yield change

Li → Li (solid)



Li→Li
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Sputtering Yield
10^1
10^2
10^3
Incident Energy (eV)
0
15
30
45
60
75
Unity sputtering
Angle of incidence (α)

Li $\rightarrow$ Li (liquid) [Y_l/Y_s]

Energy (eV)	0°	15°	30°	45°	60°	75°
10	1.19 2.10	1.10 1.29	1.07 1.23	1.02 1.10	1.00 0.98	0.99 0.88
20	1.07 1.64	1.01 1.35	1.06 1.28	1.07 1.21	1.02 1.03	0.97 0.82
50	1.10 1.45	1.08 1.33	1.06 1.25	1.03 1.18	1.01 1.07	0.96 0.82
100	1.07 1.34	1.07 1.30	1.04 1.23	1.05 1.17	1.02 1.08	0.98 0.87
200	1.06 1.30	1.06 1.26	1.05 1.21	1.02 1.14	1.02 1.08	0.99 0.96
500	1.04 1.26	1.05 1.22	1.03 1.16	1.02 1.13	1.02 1.08	1.00 1.02

Top line = 1.1/.95/1.0 profile Bottom line = 1.5/.75/1.0 profile Blue $\Rightarrow$ > 15% Yield change

Conclusions

- **Surface density layering can result in increases in the sputtering yield**
- **Density effect appears to be largest in the case of self-sputtering**
- **Further development is needed to accurately model surface layering and incorporate coordination number effects**
  - **Add additional layers to TRIM98**
  - **Vary surface binding energies**
  - **Improve statistics at low sputtering yields**
  - **Look at H recombination energy in thin layers**