Derivative-Free Optimization Methods for a Surface Structure Inverse Problem

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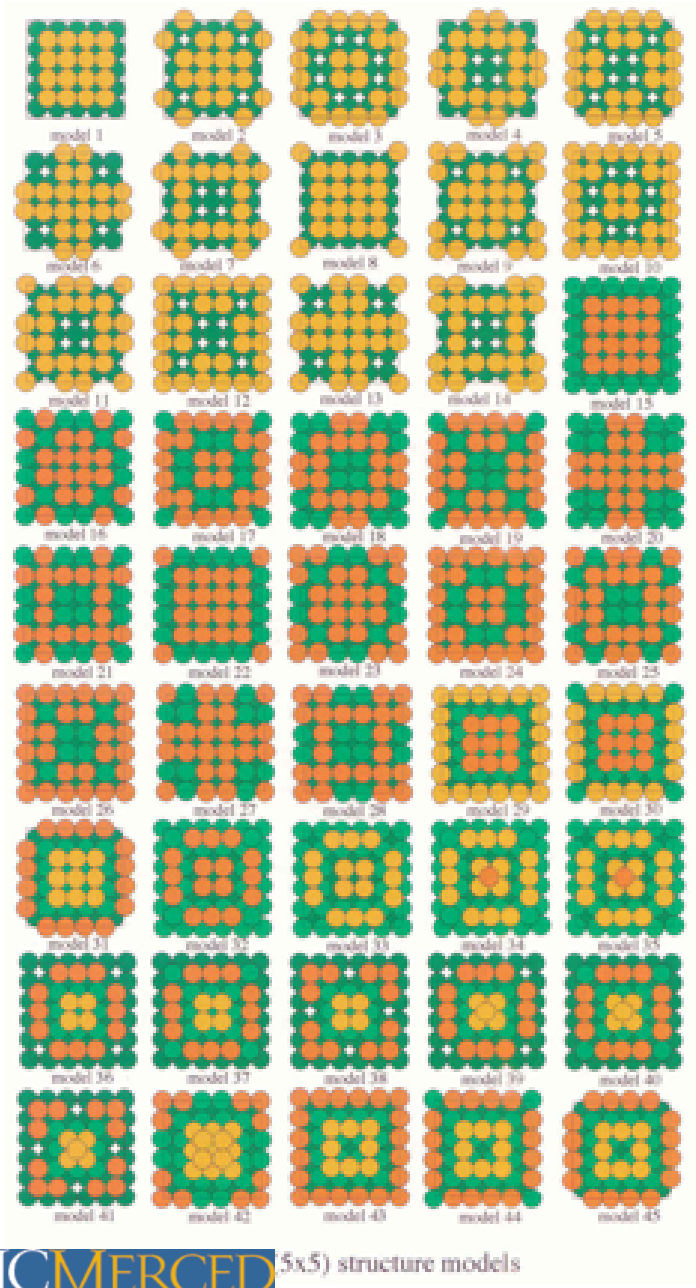
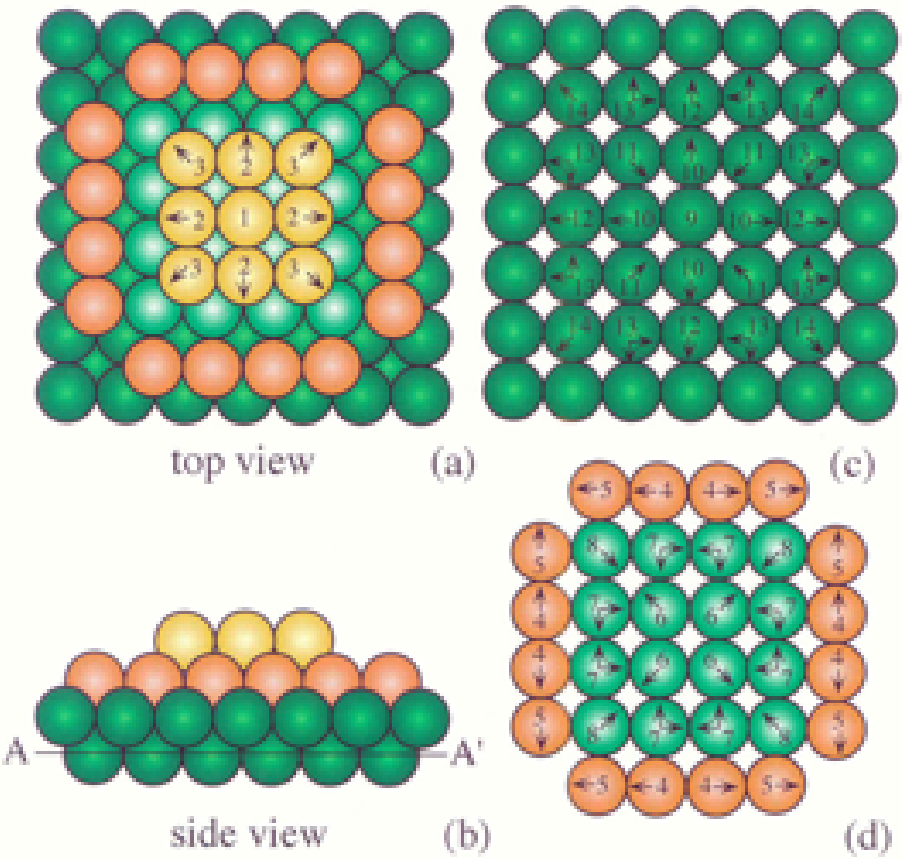
Surface structure determination

- Surface structure determines many important properties of materials.
- Possibility of electron diffraction first proposed by deBroglie (1924).
- Over 40 years before it became a tool in surface structure determination.
- Experimental data could not be quantitatively described by kinematic theory and necessitated the development of a theory of multiple scattering in the late 1960s.

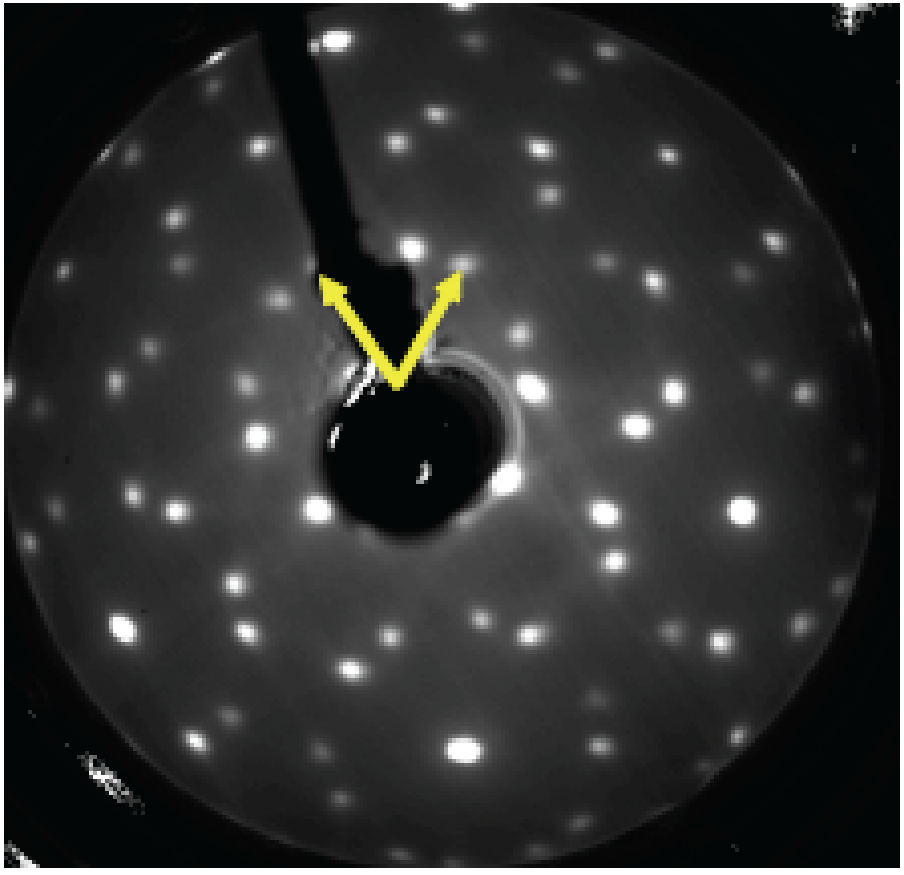
What is the correct atomic configuration?

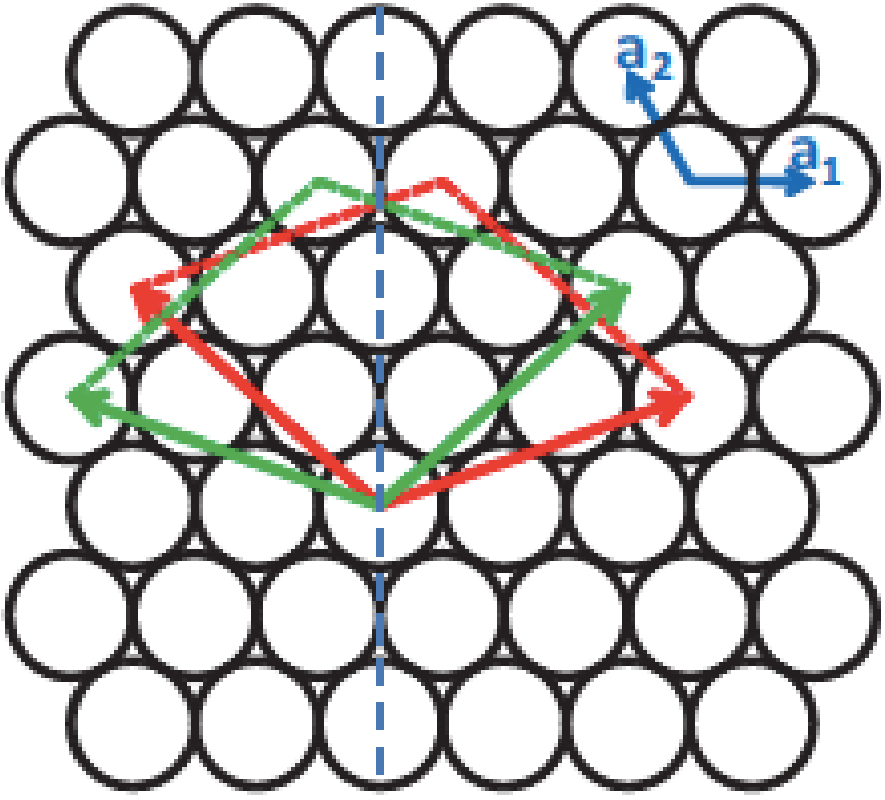


5x5) structure models



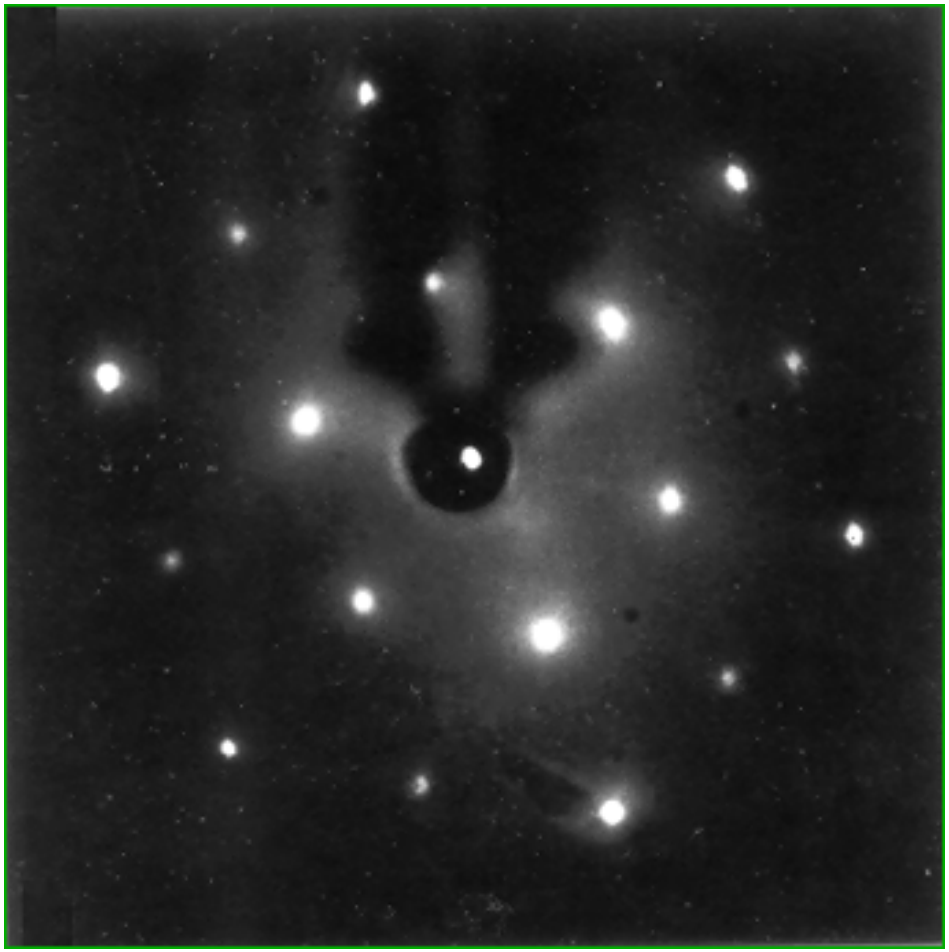
Electron Diffraction patterns (Held, G. (1974))





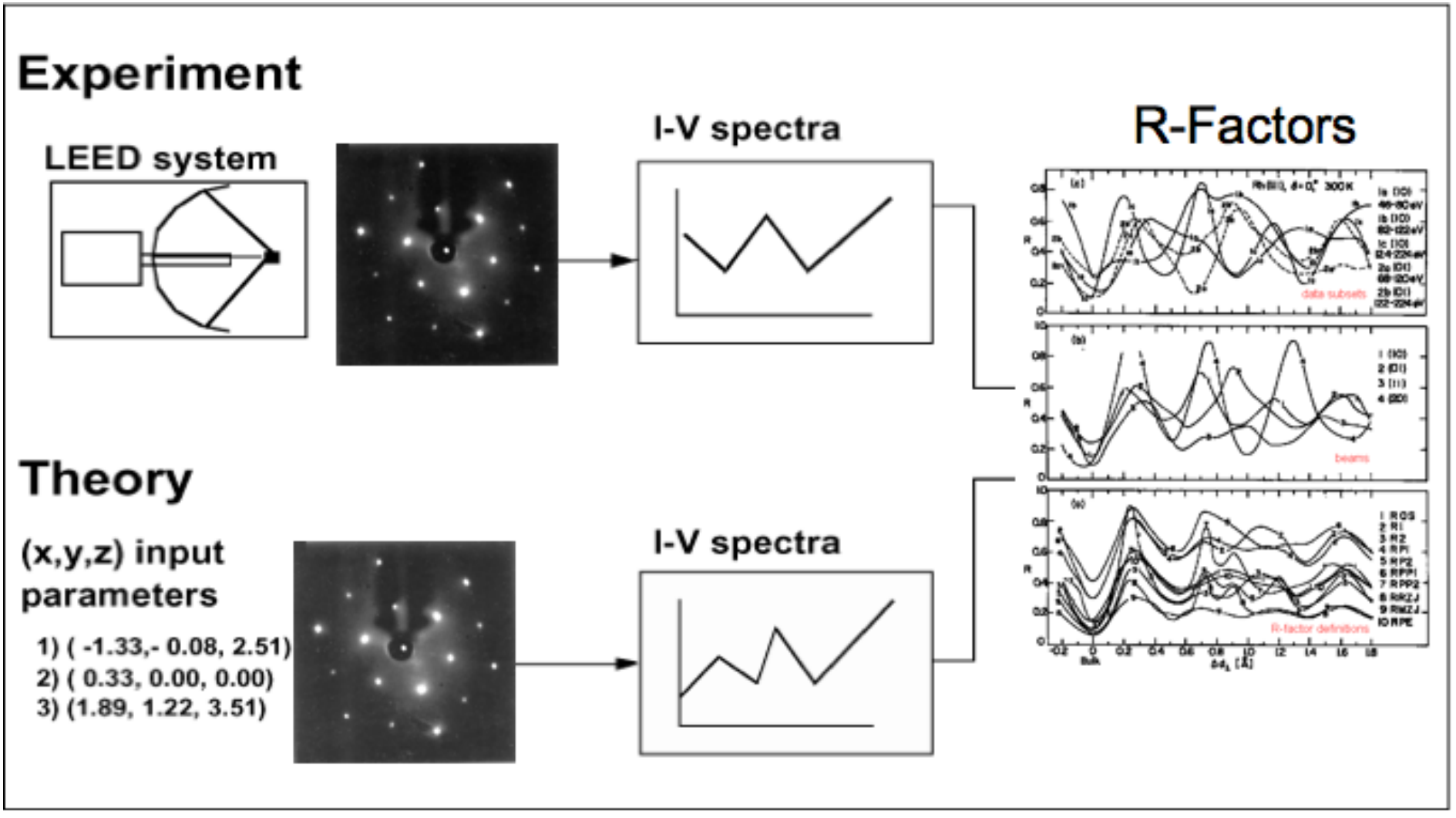
$\Theta = 0.57ML$: $p(\sqrt{7} \times \sqrt{7})\ R19°$ $\quad M = \begin{pmatrix} 3 & 1 \\ -1 & 2 \end{pmatrix}$

Low-energy electron diffraction (LEED)

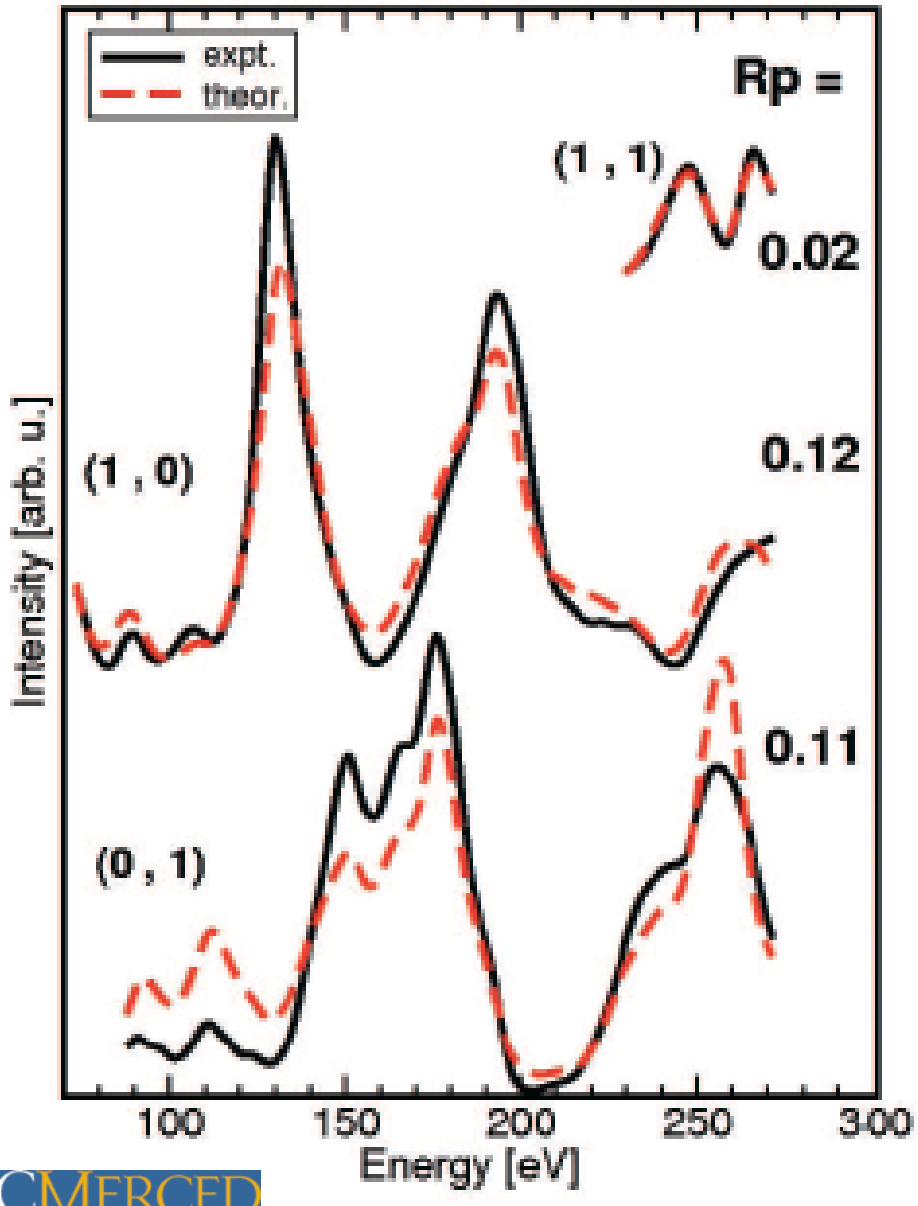


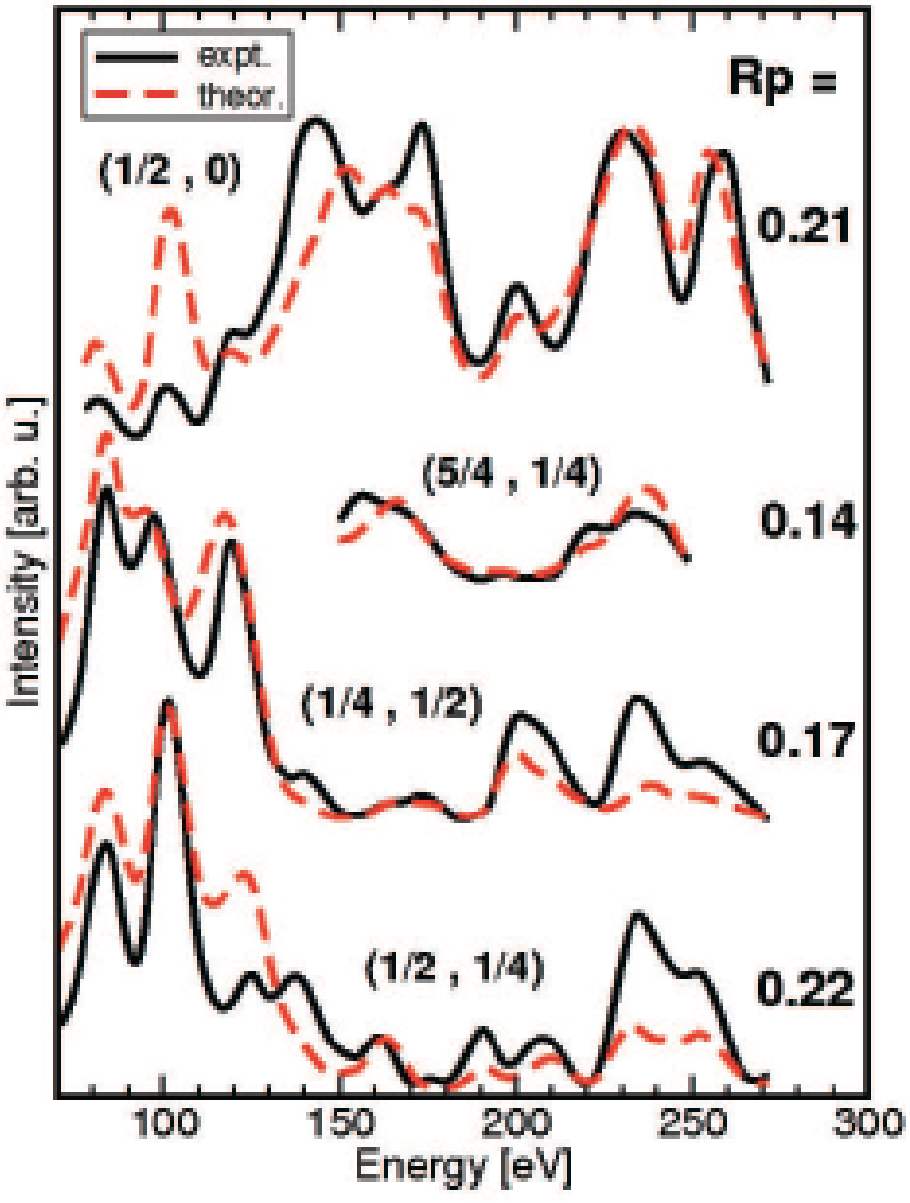
- Goal is to determine surface structure through low energy electron diffraction (LEED).
- Need to determine the coordinates and chemical identity of each atom: Li atoms on a Ni surface
- Low-energy electrons have high surface sensitive, requiring a well-ordered surface

Experimental setup



IV curves (Held, G. (1974))





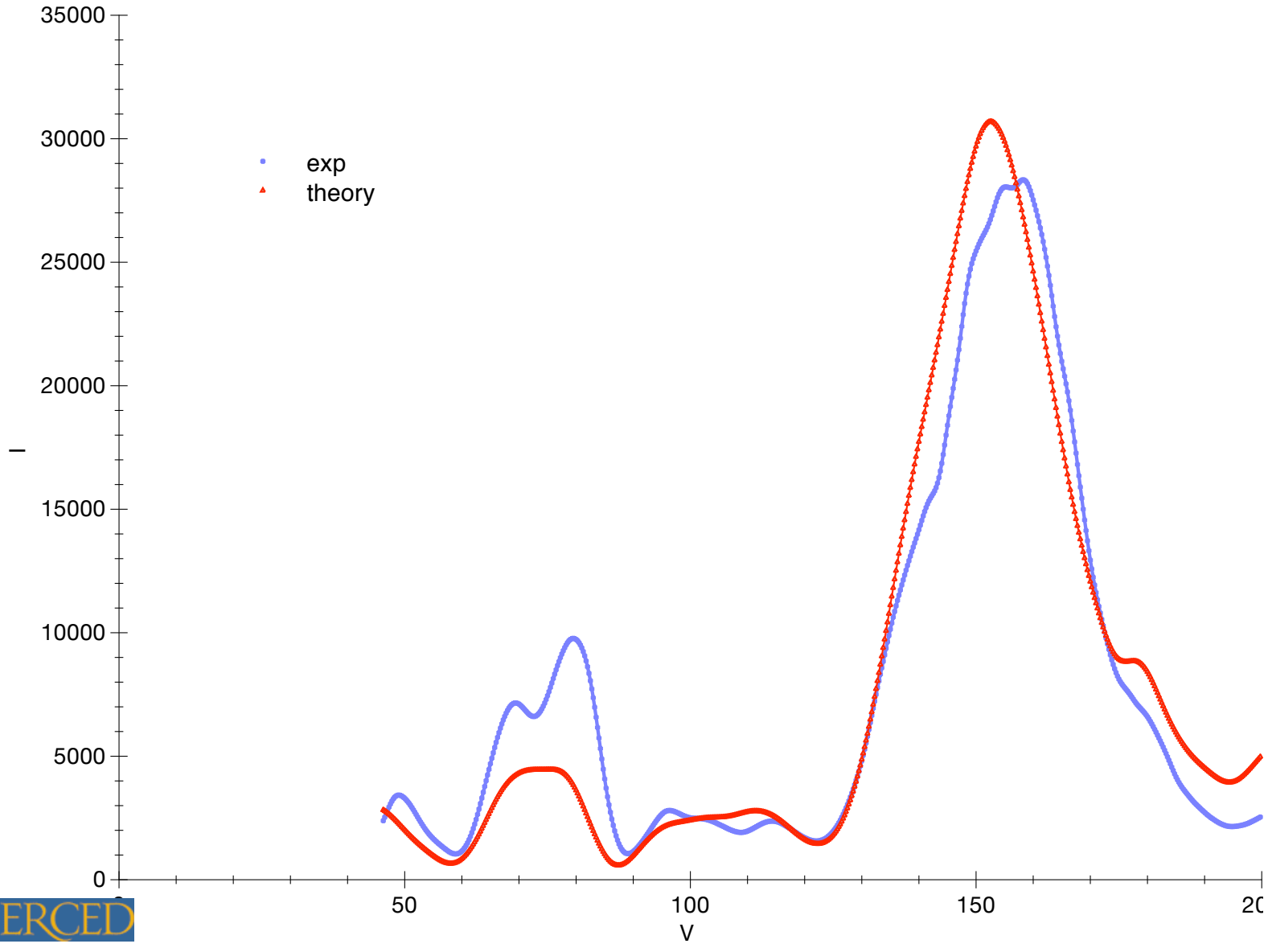
Pendry Reliability-factor (1980)

$$
\begin{aligned}
R &= \sum_{i,g}(Y_{gth} - Y_{gexp})^2 / \sum_{i,g}(Y_{gth}^2 + Y_{gexp}^2) \\
Y(E) &= L/(1 + L{V_{oi}}^2) \\
L(E) &= I'/I \\
L &\approx \sum_j \frac{-2(E - E_j)}{(E - E_j)^2 + V_{oi}^2}
\end{aligned}
$$

Pendry R-factor

- LEED curves consist for the main part of a series of Lorentzian peaks: $I \approx \sum a_j/(E - E_j)^2 + V_{oi}^2$
- Their widths are dictated by the imaginary part of the electron self-energy (optical potential): $\Delta E = 2|V_{oi}|$
- Pendry R-factor emphasizes positions of the maximum and minimum rather than the heights of the intensities
- Ideal agreement corresponds to $R = 0$; uncorrelated spectra yields $R = 1$.

Example IV curve



Optimization formulation

- Inverse problem
  - minimize R-factor, defined as the misfit between theory an experiment
  - Several ways of computing the R-factor, we will use the Pendry R-factor.
- Combination of continuous and categorical variables
  - Atomic coordinates: x, y, z
  - Chemical identity: Ni, Li
- No derivatives available; function may also be discontinuous
- Invalid (unphysical) structures lead to function being undefined in certain regions and returning special values

Previous Work

- Early attempts used Hooke-Jeeves, nonlinear least squares, genetic algorithms[1−2],
- We have also used pattern search methods
- Effective, but expensive
  - Several 1000s of function calls typically needed
  - Each function call can take several minutes on a workstation class computer

1. Global Optimization in LEED Structure Determination Using Genetic Algorithms, R. Dll and M.A. Van Hove, Surf. Sci. 355, L393-8 (1996).
2. G. S. Stone, MS dissertation, Computer Science Dept., San Francisco State University, 1998.

General MVP algorithm

Initialization: Given values Δ_0, x_0, M_0, P_0,

1 For $k = 0, \ldots,$ maxit do
2 Search: Evaluate f on a finite subset of trial points on the mesh M_k.
3 Poll: Evaluate f on the frame P_k.
4 If (successful)
5 $x_{k+1} = x_k + \Delta_k d_k$
6 Update Δ_k
7 End

- Global Search phase can include user heuristics or surrogate functions
- Local Poll phase is more rigid, but needed to ensure convergence.

Variations on LEED

- LEED
  - Multiple scattering model
  - I-V spectra computed repeatedly until best-fit structure is found
  - Computation time is proportional to the number of parameters
- TLEED (Tensor LEED)
  - For a reference structure use multiple scattering
  - Perturbation method to calculate I-V for a structure close to a reference structure
  - Efficient for local modifications (i.e. no categorical variables) - otherwise computationally expensive

Kinematic LEED as a simplified physics surrogate (SPS)

- KLEED assumes that electrons are only scattered once by the surface atoms.
- Surface unit cell size and symmetry can be determined, but not the exact positions.
- Compared to multiple scattering which takes several minutes to compute, I-V spectra from KLEED can be computed in a few seconds.

Simplified physics surrogate (SPS)

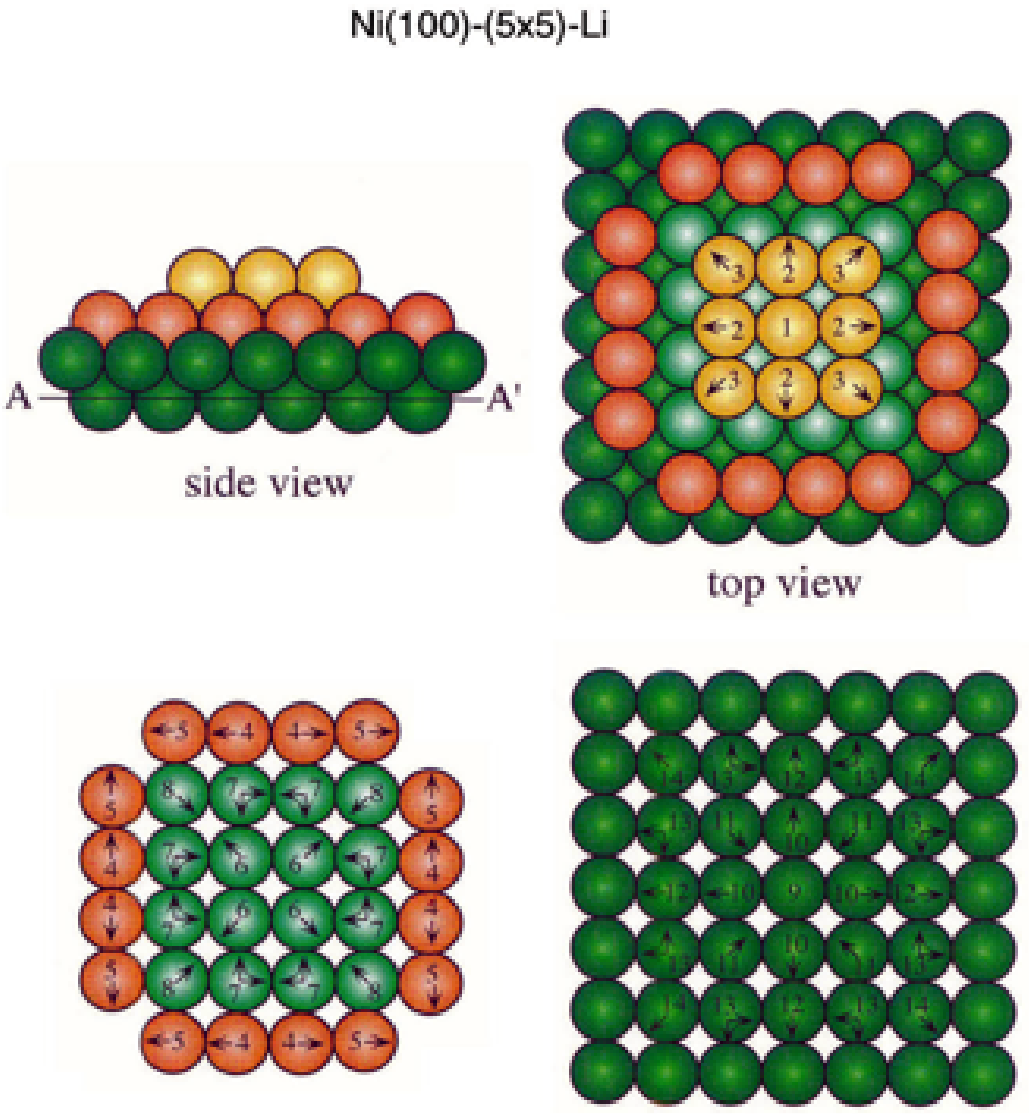
- Define $\phi_A(x) = \phi_S + \phi_I$, where

$$\begin{aligned}\phi_A &= \text{Additive surrogate,}\\ \phi_S &= \text{Simplified physics surrogate, e.g. KLEED,}\\ \phi_I &= \text{Interpolatory surrogate}\end{aligned}$$

Search:

```
1    if (first search step) {
2        Initialize φ_I with Latin Hypercube sample.
3    else {
4        recalibrate φ_I with DACE model of φ_S − f.
5        }
6    Construct φ_A = φ_S + φ_I
7    Solve min φ_A
```

Test problem

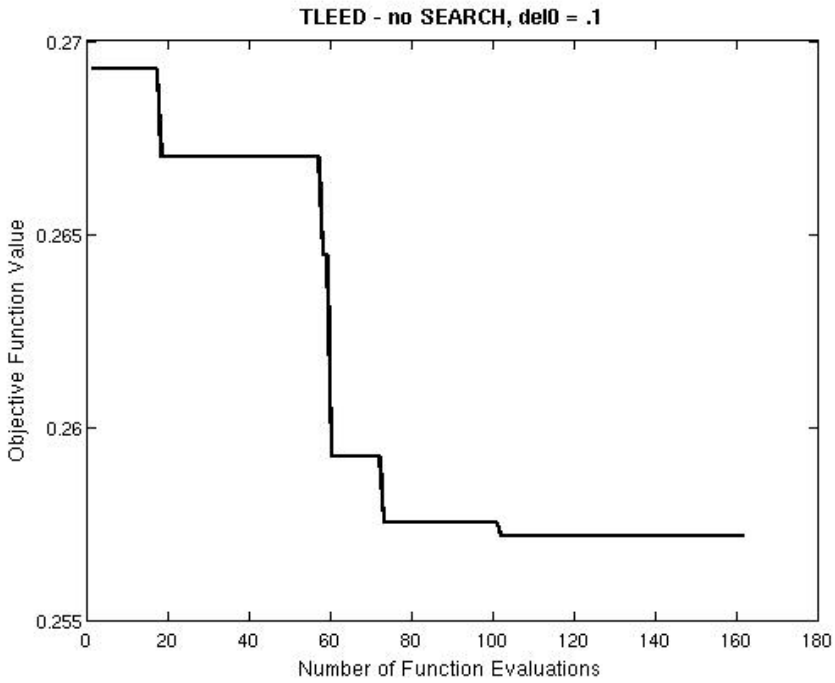


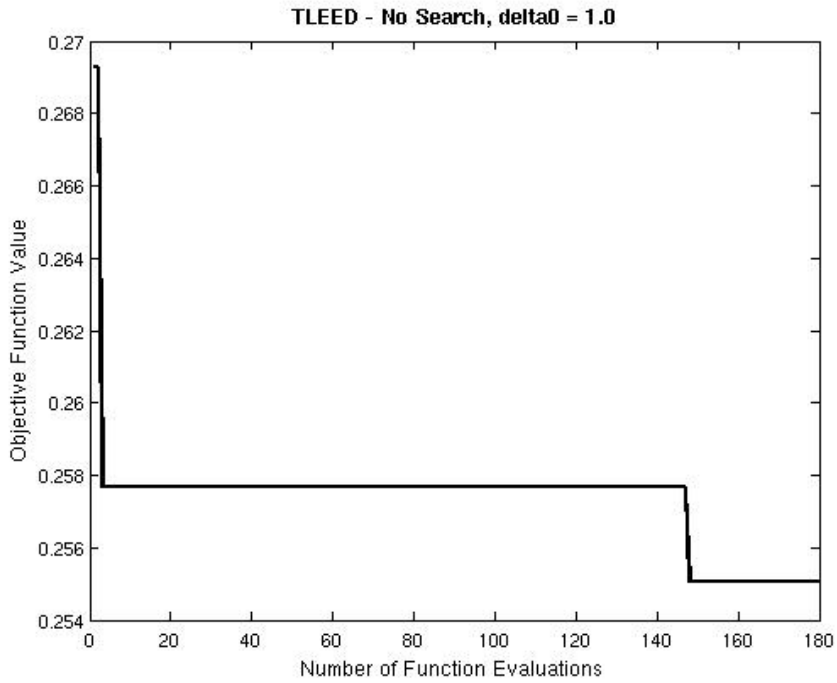
- Model 31 from set of model problem using three layers
  - 14 atoms
  - 14 categorical variables
  - 42 continuous variables
- Additional constraint added so that positions of atoms are constrained to lie within a box
- Used NOMAD (Abramson, Audet, Dennis, Le Digabel, Tribes)

Test cases

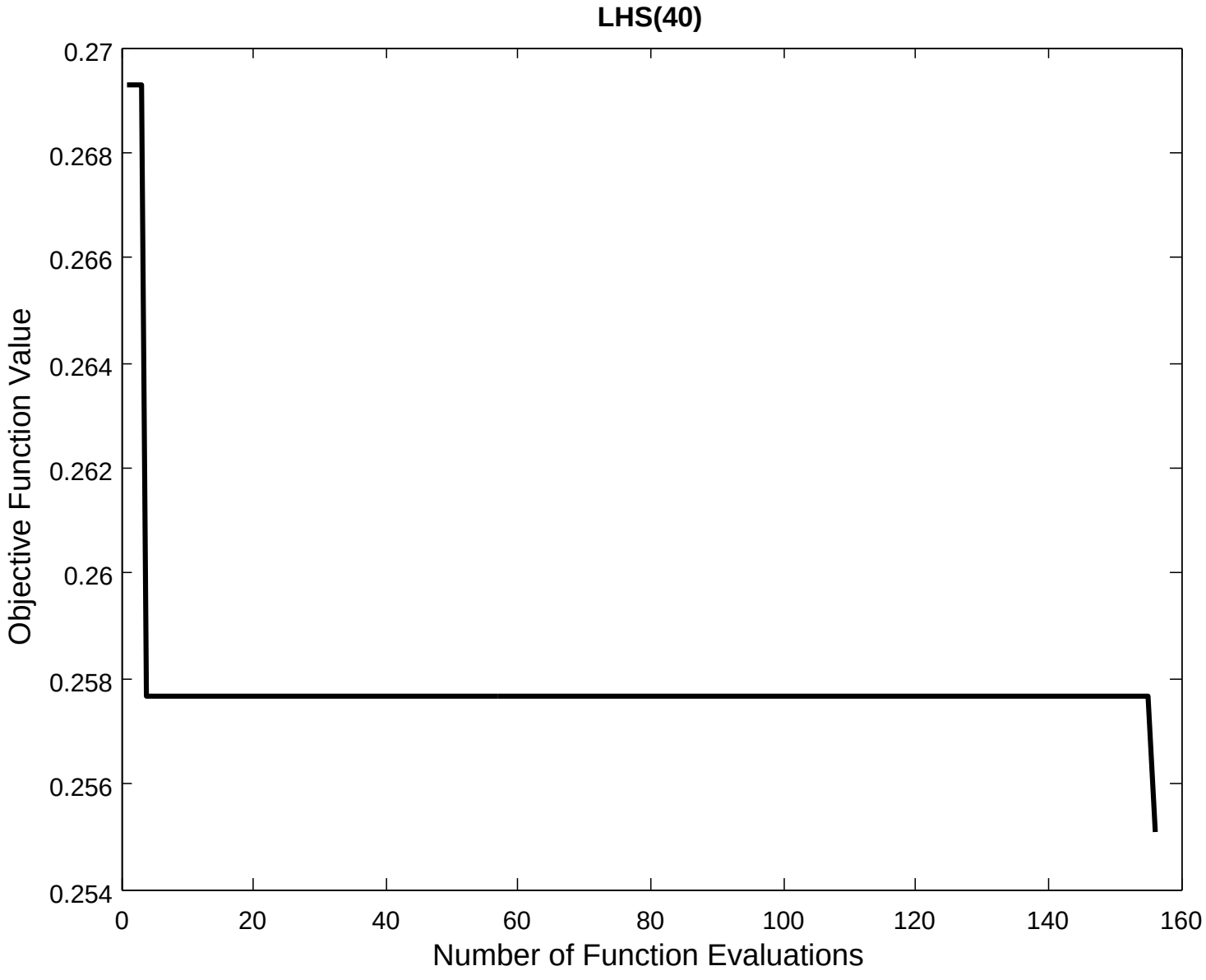
- Start with best known feasible point
- Continuous variables only
- Three different approaches
  - No search step
  - LHS search
  - Simplified physics surrogate (DACE model)

Optimization of continuous variables using no search phase

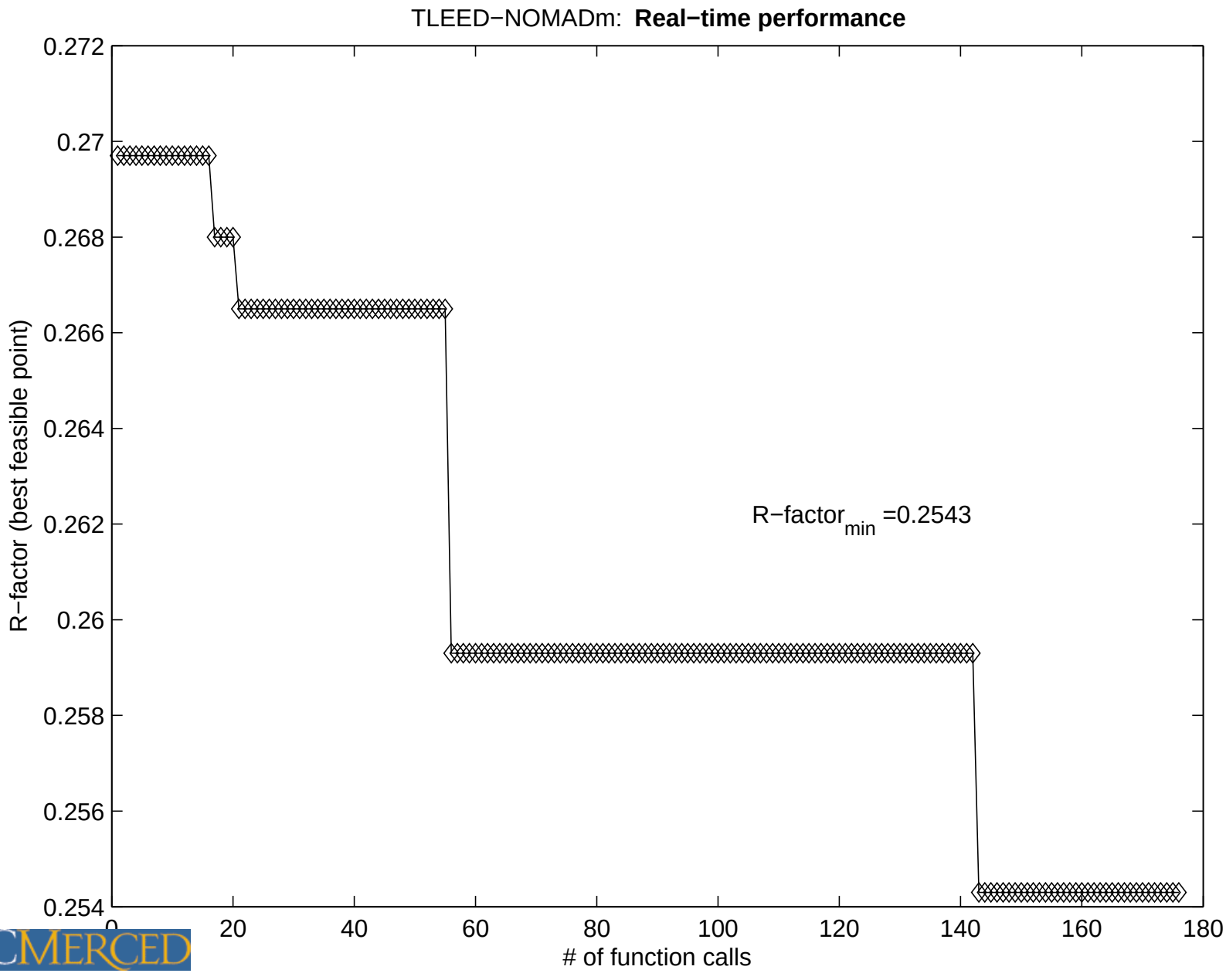




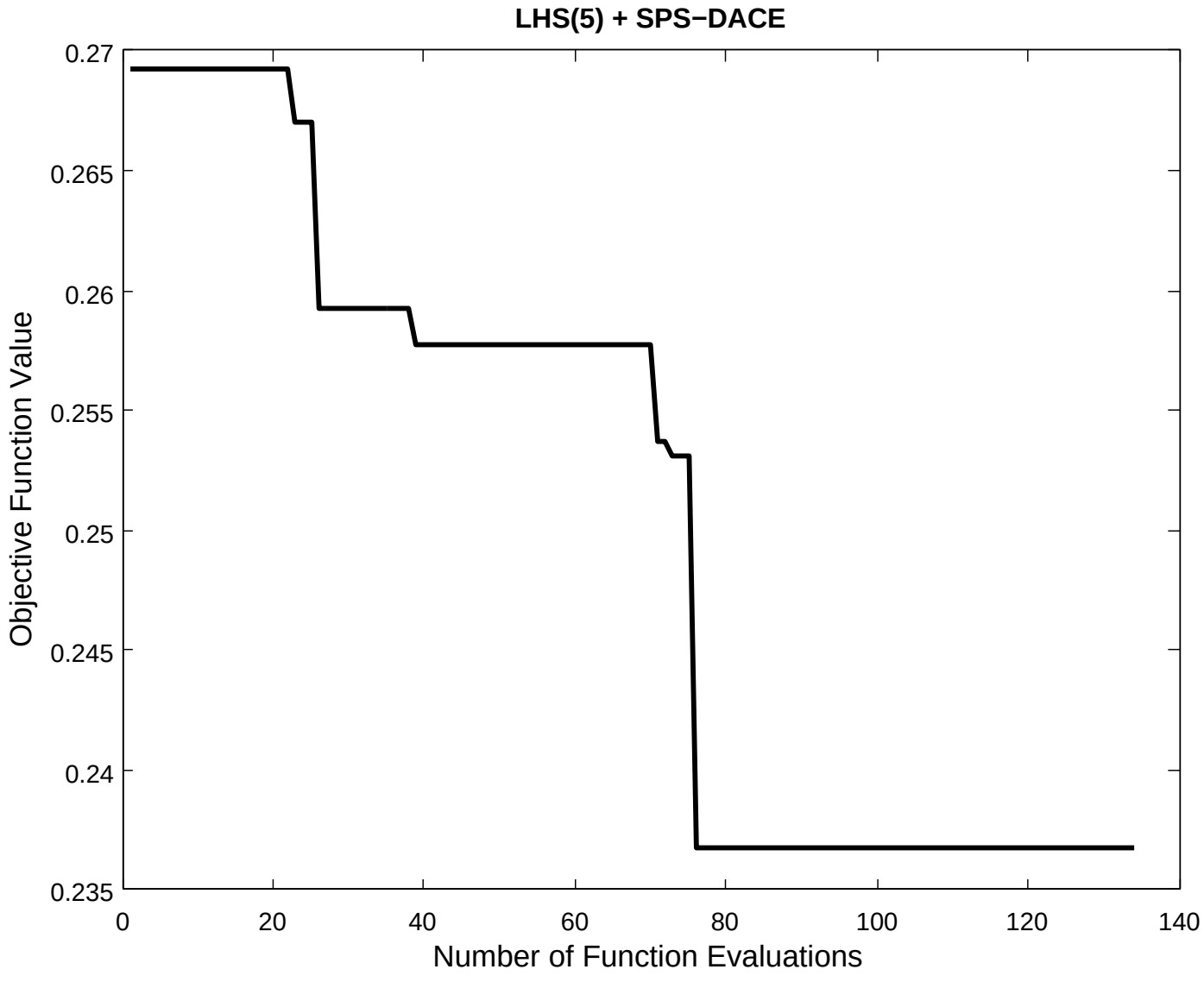
Optimization using LHS with 40 points



Optimization using additive surrogate, $\Delta_0 = 1.0$



Optimization using additive surrogate, $\Delta_0 = 0.1$



Summary of numerical results

Method	LHS	$f(x^*)$	fevals
No search	0	0.2551	180
LHS	40	0.2551	160
SPS+DACE	15	0.2543	180
SPS+DACE	5	0.2354	135

Summary

- Preliminary results indicate that performance can be enhanced by using an additive surrogate function in the search phase.
- Total number of function evaluations decreased by about 20%, which represents a reduction of ≈ 2 hours per model.
- Efficiency is (too) dependent on various algorithmic parameters.
- Need to investigate alternate interpolatory surrogates

Future Work – Chemical Identity Search