Derivative-free optimization methods for surface structure determination of nanosystems

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October 19, 2007

Abstract

Many properties of nanostructures depend on the atomic configuration at the surface. One common technique used for determining this surface structure is based on the low energy electron diffraction (LEED) method, which uses a high-fidelity physics model to compare experimental results with spectra computed via a computer simulation. While this approach is highly effective, the computational cost of the simulations can be prohibitive for large systems. In this work, we propose the use of a direct search method in conjunction with an additive surrogate. This surrogate is constructed from a combination of a simplified physics model and an interpolation that is based on the differences between the simplified physics model and the full fidelity model.

1 Introduction

Many properties of nanostructures depend on the atomic configuration at the surface. One frequently used technique for surface structure determination is the low energy electron diffraction (LEED) method [1, 2]. LEED involves the use of an electron gun to bombard a sample with a beam of electrons, with energy, and recording the diffraction pattern over a range of energies. The change in intensity (I) can then be tracked as a function of the energy (V), generating an intensity-voltage curve. Previous attempts at structure determination have employed a time intensive trial-and-error approach, to find the best-fit surface structure by comparing LEED calculations to experimental intensity spectra. Here, we propose an alternative approach based on a direct search method within a surrogate framework for solving this optimization problem.

The optimization problem is usually formulated in terms of minimizing the Pendry R-factor, which measures the misfit between theory and experiment. Several methods have been proposed for this problem including simulated simulated annealing [3], fast simulated annealing [4, 5], a modified random sampling algorithm [6] and genetic algorithms (GAs) [7]. Recently, Zhao et al. [8] applied a generalized pattern search method (GPS) to the problem of determining the structure of a

Ni(001)-(5x5)-Li surface. Pattern search methods were found to have better performance than both simulated annealing and GA, generating better trial structures with significantly fewer evaluation functions required. Nevertheless, each of the function evaluations is computationally expensive, and consequently, the total time required for a structure determination is still daunting. Here, we propose the use of an additive surrogate function for the objective function to reduce the computational effort.

2 Simplified physics surrogate

In optimization, a surrogate is an inexpensive function that replaces a more expensive one. The goal is to shift the computational effort from the true expensive functions onto the inexpensive surrogates. One fairly well understood approach for building surrogates, which has proven itself in practice [9, 10], is to interpolate a set of known data points and their function values using, for example, Kriging functions. In this work, we propose a simplified physics model or simplified physics surrogate (SPS) as a means of reducing the computational effort. The calculation of the I-V spectrum is based on a sophisticated multiple scattering model. However, all multiple-scattering methods designed for LEED scale unfavorably with the complexity of the structure being investigated, and in particular with the number of atoms in the unit cell or surface section of interest. However, in the kinematic limit, only single scattering events are included in the description of the electron diffraction. As a result, the kinematic LEED (KLEED) approximation contains significant information about the structure and we will use it as a surrogate for the multiple scattering model.

Since the KLEED approximation is not guaranteed to match the high-fidelity model even at the optimal solution, we employ an additive surrogate composed of an interpolatory function in addition to the simplified physics surrogate. The interpolatory part of the surrogate is designed to fit the difference between the multiple scattering LEED function value and the simplified physics model. For simplicity, we will denote the multiple scattering LEED function values as the "true" values. Let $\phi_A = \phi_S + \phi_I$, where ϕ_A = additive surrogate, ϕ_S = simplified physics surrogate, e.g. KLEED, and ϕ_I = interpolatory surrogate, e.g. DACE model. The surrogate optimization and update is performed as follows. First, we initialize the additive surrogate by computing a set of function values at a set of points generated by a Latin Hypercube sample (LHS). The simplified model function ϕ_S is also evaluated at the LHS data points. Once the true and surrogate function values are known for the initial data points, we estimate ϕ_I, by interpolating the difference between the two. In our approach, the interpolation is performed using the DACE [11] MATLAB kriging toolbox.

3 Numerical Results

In this work, we employed the NOMADm software package [12], which is a MATLAB implementation of a mesh adaptive direct search (MADS) algorithm [13]. Each iteration of MADS contains two steps – a SEARCH step and a POLL step. In the SEARCH, we construct and quickly optimize the inexpensive additive surrogate to generate good trial points for the true function to evaluate. If any of these trial points gives a reduced function value (as measured by the true function) the iteration is

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

Figure 1: Comparison of various search strategies

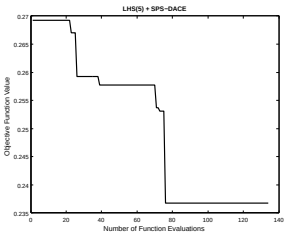

Figure 2: Optimization using additive surrogate, $\Delta_0 = 0.1$

declared successful. Otherwise, the POLL step is performed and the algorithm proceeds as normal. After each iteration, the additive surrogate is updated with the newly evaluated points and their true and SPS function values.

We tested our additive surrogate on a test problem based on the complex Ni(001)-(5x5)-Li surface, formed by adding lithium (Li) atoms onto a (001) nickel (Ni) crystal (5x5) superlattice surface. Due to symmetry, there are only 14 atoms on this surface, resulting in 42 (14×3) optimization variables. We compared our results against the numerical experiments performed by Zhao *et al.* [8]. Table 1 displays the results for using the additive surrogate versus no search and a simple LHS search, in which columns 2-4 contain the number of initial LHS points used, the best objective function value $f(x^*)$ found, and the number of true function evaluations (fevals). The results indicate a reduction in the number of true function evaluations by as many as 45, which translates into approximately 2 hours of CPU time per optimization run. Figure 2 contains the iteration history for the last row in Table 1.

4 Conclusions

Our results indicate that performance can be enhanced by using an additive surrogate function in the SEARCH phase of a pattern search method. The total number of function evaluations decreased by approximately 20%, which represents a reduction of ≈ 2 CPU hours per optimization run. In addition, the case using SPS with DACE and an initial LHS sample size of 5 was able to achieve a better solution than any of the previous results. The efficiency can be highly dependent on certain algorithmic parameters though, and further research is necessary to understand the effect of the DACE model on performance.

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