Microscopic/Stochastic Timesteppers and "Coarse" Control: a KMC Example

C. I. Siettos, A. Armaou, A. G. Makeev, and I. G. Kevrekidis
Dept. of Chemical Engineering, Princeton University, Princeton, NJ 08544

Mathematical models, whether identified on-line during an experiment or derived from first principles, constitute the backbone of modern control practice. Models of chemical and transport processes (material, energy, and momentum balances) traditionally take the form of deterministic ordinary or partial differential/algebraic evolution equations for macroscopic variables (such as concentrations). An arsenal of mathematical and computational tools targeted at such macroscopic models has been developed over the years for the performance of macroscopic, system-level tasks such as temporal simulation, stability and bifurcation analysis, optimization, design and control. For many processes of current interest, however, the best available description of the physics (through molecular dynamics (MD), kinetic Monte Carlo (KMC), kinetic theory based Lattice-Boltzmann (LB), or Markov chain simulators) operates at a vastly different scale from that at which the questions of interest are asked and the answers are required (such as operating diagrams and controller design for expected reaction rates). The implication is that macroscopic rules (description at a higher level) can somehow be deduced from microscopic ones (description at a much finer level). In most current problems, however, ranging from ecology to materials science and from chemistry to engineering, the closures required to translate microscopic/stochastic models to a high-level, macroscopic description are simply not available. A so-called "coarse timestepper" approach to stability and bifurcation calculations has recently had some success in circumventing the derivation (through explicit closures) of macroscopic descriptions (Theodoropoulos et al., 2000; Gear et al., 2002; Makeev et al., 2002; Runborg et al., 2002). Using short "bursts" of appropriately initialized and processed microscopic simulations coupled with system identification, one tries to analyze the behavior of macroscopic equations *without ever obtaining these equations in closed form.*

The purpose of this article is to suggest that such "coarse timesteppers" have the potential to become a bridge between microscopic/stochastic modeling and well-established control design techniques, such as linear feedback or model predictive control. The key assumption is that deterministic, macroscopic, *coarse* models exist *and close* for the expected behavior of a few low *moments* of microscopically evolving distributions (such as for surface coverages, the zeroth moments of adspecies distributions on a lattice); however, they are unavailable in closed form. Computer-assisted location of steady states or design of simple stabilizing controllers requires the *evaluation* of certain quantities from these unavailable equations (residuals, Jacobians, Hessians, and so on). One formulates a framework in which it is possible to computationally *identify* these quantities "*on demand*" [or "*just in time*" (Cybenko, 1996)] from judicious microscopic simulations, since, lacking the explicit equation, we cannot simply evaluate them. We then pass them to the algorithm that performs the macroscopic task (steady state location, control design algorithm). In effect, through a computational superstructure built around the microscopic simulation code, we "fool" the macroscopic numerical algorithms into performing tasks for an equation that is not explicitly available.

We start with a brief overview of the "coarse timestepper" [detailed discussions can be found in Gear et al. (2002) and Makeev et al. (2002)], and a simple version of the framework for macroscopic controller design based on microscopic simulators. Our illustrative example is a kinetic Monte Carlo (KMC) simulation of a simple surface reaction scheme. We first obtain its time-stepper-based coarse bifurcation diagram. We then demonstrate the results of implementing a simple *coarse controller* for stabilizing an open-loop unstable coarse (macroscopic) equilibrium of the KMC model, and conclude with a short discussion.

Coarse Controllers for Microscopic Simulators

The following steps outline a simple framework for designing coarse, macroscopic controllers, based on information extracted at the system level from microscopic models, sidestepping the derivation of a closed form macroscopic description:

Coarse timestepper

• Choose appropriate statistics $\boldsymbol{x}$ (typically zeroth- or first-order moments of evolving probability densities $\boldsymbol{X}$) that

Correspondence concerning this article should be addressed to I. G. Kevrekidis.
Permanent address of A. G. Makeev: Faculty of Computational Mathematics and Cybernetics, Moscow State University, Moscow, 119899, Russia.

we believe can deterministically describe the long-term *macroscopic* behavior of the system under study. This implies that higher-order statistics become quickly (say, over a few collision times in a microscopic simulation) slaved to these lower ones (they evolve to functionals of the lower ones, alternatively evolve towards a "slow manifold" parameterized by the lower ones). These choices also determine a *restriction* operator M, mapping (projecting) the microscopic-level description $\boldsymbol{X}$, into the macroscopic description: $\boldsymbol{x} = M\boldsymbol{X}$;

- Choose an appropriate (nonunique!) *lifting* operator μ, mapping the macroscopic description $\boldsymbol{x}$ to one or more consistent microscopic descriptions $\boldsymbol{X}$; that is, construct distribution(s) consistent with (*conditioned on*) a few low-order moments [see Gear et al. (2002) and Makeev et al. (2002) for examples and discussion]. Lifting from the microscopic to the macroscopic and then restricting again should have no effect, that is, $M\mu = I$ (except roundoff)
- Prescribe a macroscopic initial condition (such as concentration profile, coverage) $\boldsymbol{x}(t_0)$
- Transform it through lifting to one (or more) consistent microscopic realizations $\boldsymbol{X}(t_0) = \mu\boldsymbol{x}(t_0)$
- Evolve this(ese) realization(s) using the microscopic simulator for the desired short macroscopic time T, generating the value(s) $\boldsymbol{X}(T)$. The choice of T is associated with the (estimated) spectral gap of the linearization of the unavailable macroscopic equation [see Gear et al. (2002) and Makeev et al. (2002) for discussion]
- Obtain the restrictions $\boldsymbol{x}(T) = M\boldsymbol{X}(T)$.

The procedure is a "black box" "coarse timestepper" $\Phi_T(x_0) = \boldsymbol{x}(T)$ with $\boldsymbol{x}_0$ as initial condition.

A coarse stability/bifurcation framework

A coarse stability/bifurcation framework can now be implemented as a computational superstructure, a "shell," around repeated calls to this coarse timestepper. *Coarse (macroscopic) steady states* can be obtained as fixed points of the iteration $\boldsymbol{x}(k+1) = \boldsymbol{\Phi}_T[\boldsymbol{x}(k)]$. These are *not* steady states of the (stochastic, constantly varying) KMC simulation; they are "almost always" steady states of the (expected values of the) low moments $\boldsymbol{x}$ of the KMC-evolved distributions $\boldsymbol{X}$. The action of the (slow) local Jacobian of the mapping $\boldsymbol{D\Phi}$ is estimated "on demand" using numerical derivatives and "lift-run-restrict" evaluation of $\boldsymbol{\Phi}$. For large, discretized distributed coarse systems, calls to the timestepper with ϵ-nearby coarse initial conditions are used to estimate the requisite matrix-vector products $\boldsymbol{D\Phi}\cdot\epsilon$. Contraction mappings like Newton-Raphson, or so-called Newton-Picard timestepper based algorithms using Krylov subspace iterations [such as the Recursive Projection Method of Shroff and Keller, (1993)], are used to find the coarse steady states [see also Lust (1997) and Tuckerman and Barkley (2000)].

Coarse input-output data, especially in the neighborhood of the located macroscopic steady state, can be used to perform additional coarse (such as nonlinear ARMAX) model identification; extended Kalman filters, extended least squares (ELS), or recursive maximum likelihood (ML) algorithms (Åstrom and Wittenmark, 1995; Ljung, 1999) can be used for the estimation of the polynomial coefficients. The important element is that the initial conditions of the coarse time-stepper (as opposed to those of a physical experiment) can be repeatedly set *at will*. We can now proceed to the next step.

Coarse control framework

The above computational framework serves as an "on demand" identification methodology for righthand sides, "coarse slow" Jacobians, coarse derivatives with respect to parameters, and so on; in short, of precisely the quantities that a control design algorithm would need to be evaluated from a macroscopic model, had such a model been available, to per-

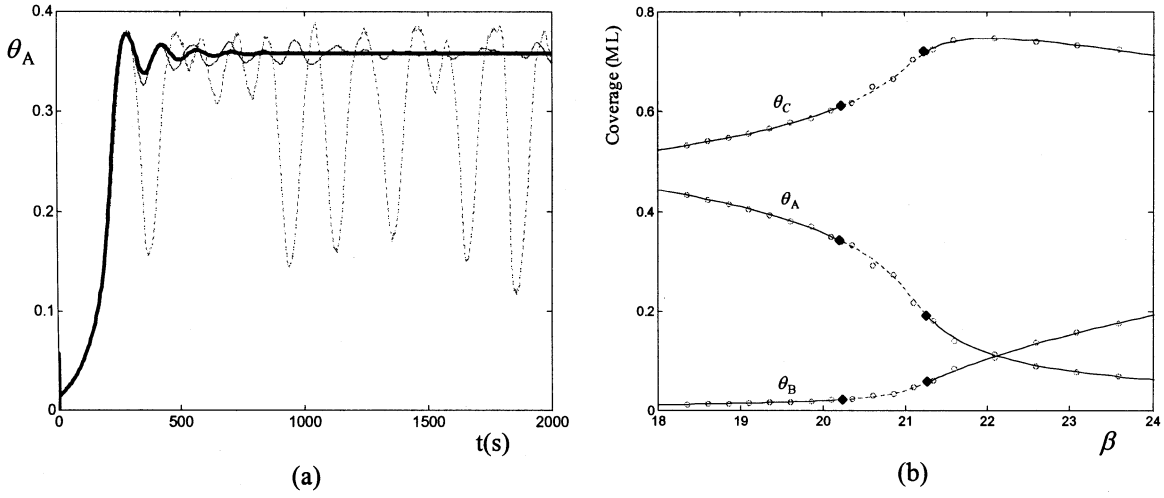

Figure 1. (a) Open-loop θ_A response (KMC simulator, $\beta \approx 20$ and $\mu = 0.36$, system sizes $N = 200^2$ (dotted line) and $N = 1{,}000^2$ (thin solid line), and single realization ($N_{run} = 1$); deterministic response (thick solid line) is for comparison. (b) one-parameter coarse KMC bifurcation diagram (derived using $N = 200^2$ and $N_{run} = 2{,}000$).

Two supercritical coarse Hopf bifurcation points are marked with diamonds; circles correspond to the coarse KMC steady states. Mean field bifurcation diagram (for comparison); stable (unstable) steady states are represented by solid (dotted) lines.

form its task. We simply substitute the macroscopic function evaluations with this computational shell around the microscopic timestepper. Using the concepts of separation and certainty equivalence principles, we then perform control tasks, such as the design of local linear stabilizing controllers. Optimal controller and model predictive controller design, even though considerably more intricate, conceptually follow directly in a discrete-time, coarse timestepper based framework.

Simple Kinetic Monte Carlo Model for Catalytic CO Oxidation. Our illustrative model is a "stochastic simulation algorithm" (Gillespie, 1976, 1977) Monte Carlo realization of a simplification of the kinetics of catalytic CO oxidation whose mean field description is given by the following equations

$$\frac{d\theta_A}{dt} = a\theta^* - \gamma\theta_A - 4k_r\theta_A\theta_B \tag{1a}$$

$$\frac{d\theta_B}{dt} = 2\beta\theta^{*2} - 4k_r\theta_A\theta_B \tag{1b}$$

$$\frac{d\theta_C}{dt} = \mu\theta^* - \eta\theta_C \tag{1c}$$

where θ_i represent the coverages of species (i = A,B,C, respectively, CO, O and an inert species C) on the catalytic surface; $\theta^* = 1 - \theta_A - \theta_B - \theta_C$; $\alpha,\beta,\gamma,\mu,\eta$ are associated with CO adsorption, O dissociative adsorption, CO desorption, and C ads/desorption rates; and k_r is the reaction rate constant. Simulation results were obtained for $\alpha = 1.6$, $\gamma = 0.04$ and $k_r = 1$, $\eta = 0.016$. The bifurcation parameter was β and the control variable was μ (physically varied through gas phase pressures of O_2 and the inert C, respectively). The KMC simulations approximate the solution of the corresponding master equation, which describes the evolution of the probability (PDF) of finding the system in a certain configuration (Makeev et al., 2002). Figure 1a shows the open-loop behavior (at $\beta \approx 20$ and $\mu = 0.36$) of θ_A using the KMC simulator for two system sizes $N = 200^2$ and $N = 1{,}000^2$, and a single realization $N_{\text{run}} = 1$; the deterministic response is also given for comparison purposes. Figure 1b shows a one-parameter bifurcation diagram (both mean-field and the coarse KMC timestepper one) with respect to β. The steady states on the coarse bifurcation diagram have been obtained as fixed points of the microscopic timestepper for $N = 200^2$ and $N_{\text{run}} = 2{,}000$ through the procedure and have the correct "coarse stability." The model exhibits two supercritical Hopf bifurcations (at $\beta \approx 20.3$ and $\beta \approx 21.2$) and stable oscillatory behavior in-between.

We want a stabilizing controller for the macroscopic (expected) unstable steady state at $\beta \approx 20.7$. This coarse steady state (evaluated through the $T = 0.025$ coarse timestepper with $N = 2{,}000^2$ and $N_{\text{run}} = 10$, upon convergence of the Newton-Raphson to a residual of $O(10^{-4})$ for $\epsilon \sim 10^{-3}$) is $\theta_A \approx 0.290937$, $\theta_B \approx 0.029468$, $\theta_C \approx 0.651079$. At stationarity, the estimates of the Jacobian $\boldsymbol{A}$ and control matrix $\boldsymbol{B}$ of the unknown macroscopic equation are

$$\boldsymbol{A} \approx \begin{bmatrix} 0.95851 & -0.06420 & -0.0313625 \\ -0.058825 & 0.91360 & -0.0559 \\ -0.0985 & -0.0064625 & 0.99324 \end{bmatrix}$$

$$\boldsymbol{B} \approx \begin{bmatrix} -1.5E-05 \\ 4.9625E-04 \\ 3.3755E-04 \end{bmatrix}$$

(This coarse steady state is estimated within 1% of the mean field one; the Jacobian and control matrix elements are also well estimated.) These matrices, which would appear in a standard discrete time local linear stochastic state space model of the type $\boldsymbol{x}(k+1) = \boldsymbol{A}\boldsymbol{x}(k) + \boldsymbol{B}\boldsymbol{u}(k) + \boldsymbol{w}(k)$, (where the vector $\boldsymbol{x} \in \boldsymbol{R}^n$ represents the state variables; $\boldsymbol{u} \in \boldsymbol{R}^p$ is the control vector of the system while the vector $\boldsymbol{w} \in \boldsymbol{R}^n$ denotes the process noise), are a byproduct of the fixed point/continuation algorithm for a coarse steady-state location. For large-size problems (such as those arising in discretized coarse PDEs), RPM-type algorithms will identify the "coarse slow" Jacobian (for dissipative PDEs in which a sepa-

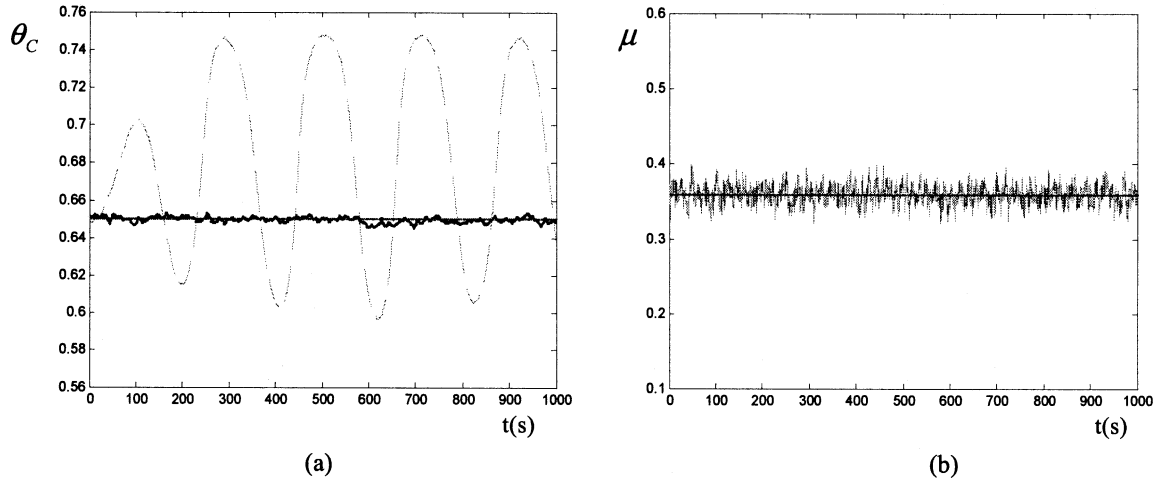

Figure 2. (a) Open-loop (dashed line) and closed-loop (solid line) θ_C response (KMC simulator, $N = 200^2$, $N_{\text{run}} = 10$); (b) corresponding closed-loop response of the manipulated variable μ (solid line); dotted lines correspond to nominal steady-state conditions.

ration of "coarse slow" and "coarse fast" time scales arises naturally). Such estimates can be made more accurate using the coarse timestepper as an experiment, repeatedly initializing it at will in the neighborhood of the coarse steady state. Discrete time, stochastic state-space models of the standard form above, along with Gaussian uncorrelated noise sequence assumptions for $\boldsymbol{w}(k)$, bring textbook linear identification theory to bear on the problem; colored noise or even nonlinear identification tools may be progressively incorporated.

For our linear feedback controller design, we consider the case of high process noise, dictating an observer for the estimation $\hat{\boldsymbol{x}}$ of the deviation state variables from the coarse steady state. The controller thus takes the form of a linear feedback $\mu - 0.36 \equiv u = -\boldsymbol{K}\hat{\boldsymbol{x}}$ where the gain matrix $\boldsymbol{K}$ is calculated on the basis of the separation principle. The simple illustrative choice here is to use a discrete Kalman filter (Kalman and Bucy, 1961, Sage and White, 1977) as an observer, and pole placement (Kailath, 1980) for the controller design. The eigenvalues of the *discrete time* open-loop system as calculated from the estimated coarse Jacobian are $\lambda_1 = 0.865137$, $\lambda_{2,3} = 1.000106 \pm 0.00565\mathrm{i}$, consistent with instability and oscillatory behavior. The noise covariance matrix, with covariance $\mathrm{Cov}[\boldsymbol{w}(k), \boldsymbol{w}(j)] = \boldsymbol{W}\delta(k-j)$, required for Kalman filter design, was obtained numerically from the microscopic simulator. To stabilize the unstable coarse steady state, we placed the coarse eigenvalues to: $\lambda_1 = 0.865137$, $\lambda_2 = 0.98$, and $\lambda_3 = 0.99$. The required gains were $k_1 \approx 15$, $k_2 \approx -24.35$, $k_3 \approx 12.74$. The result of these choices is shown in Figure 2a, comparing open-loop and closed-loop responses of θ_C for the KMC model with $N = 200^2$, and $N_{\mathrm{run}} = 10$. If the feedback control law is to be applied to a larger system, one should, in general, test that the size of the simulated system is representative,

One can claim that this simple control scheme, using a discrete Kalman filter as an observer, locally stabilizes the unstable coarse steady state of the (expected value of the) system. Figure 2b shows the corresponding dynamics of the control variable. Filtering of the noisy simulation data can be helpful (Lou and Christofides, 2002); here, we chose a third-order time-varying FIR filter (Savitzky and Golay, 1964) in a window frame of 41 sample points. The smoothened values $\boldsymbol{x}_s$ are then used on-line to implement a linear feedback control law of the form $\mu - 0.36 \equiv u = -\boldsymbol{K}\boldsymbol{x}_s$. A more detailed view of the corresponding smoothened responses of the controlled and the manipulated variable is given in Figure 3. We demonstrate the performance of the "*off-line*" *designed* coarse controller using the KMC simulator as the experiment. To implement real-time control methodologies that use the KMC timestepper as a reference model (such as real-time model predictive control), the microscopic simulation time has to be comparable to the real-time evolution of the process. We have been developing schemes to facilitate this (such as "*the gaptooth scheme*" (Gear et al., 2002; Kevrekidis et al., 2003)).

Under comparable considerations, we have promising results (Rico-Martinez et al., 2003; Siettos et al., 2002) for using coarse control as a part of an adaptive scheme that enables microscopic simulators (or experiments) to automatically trace coarse bifurcation diagrams and to converge to low codimension coarse bifurcation points. Variance reduction plays a vital role in all these tasks.

Discussion

The proposed framework aims at establishing a synergism between "conventional" control design techniques on the one hand, and microscopic complex systems modeling on the other. Let us make clear that this procedure can only be useful if a macroscopic description is *conceptually possible*, yet unavailable in closed form; if accurate macroscopic models *are* available in closed form, one, of course, should use them for designing model-based controllers directly. What is proposed here is a systematic way of designing controllers for microscopic models based on the information that one would obtain from macroscopic models, had these models been available in closed form. *Conceptual* extensions to coarse time-stepper based model-predictive control, (local) feedback-linearization, optimal control, and "coarse optimization"

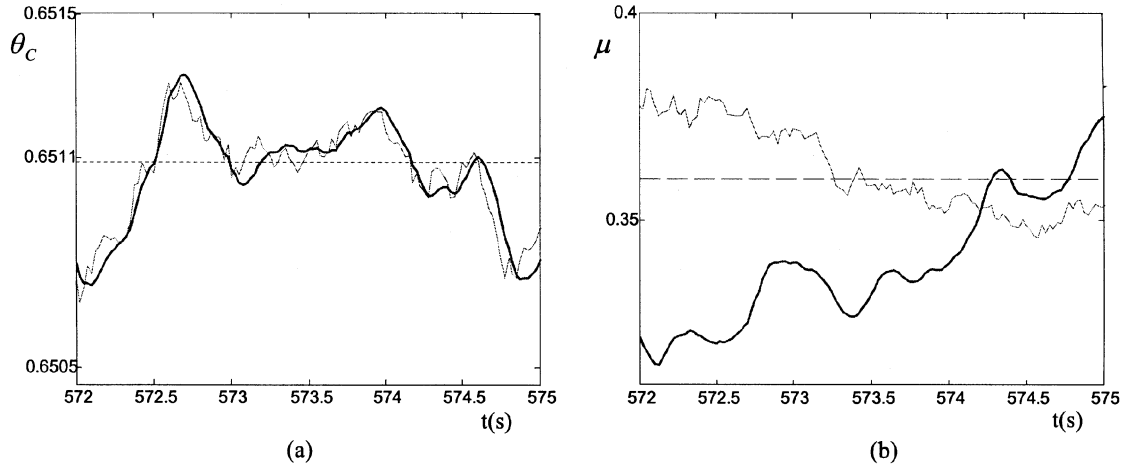

Figure 3. Detailed view of closed-loop responses of (a) controlled variable θ_C and (b) manipulated variable μ.

Thin solid lines correspond to the responses obtained without the smoothing filter, thick solid lines correspond to the responses by implementing the smoothing filter, while dotted lines correspond to nominal steady-state conditions.

appear straightforward, although there will be many nontrivial theoretical and implementation problems. System based techniques (identification) and ideas (separation of time scales between "governing" and "slaved" moments for the microscopic dynamics, as well as between "slow coarse" and "fast coarse" modes in dissipative PDEs) lie at the core of this "computational enabling technology" that bridges microscopic timesteppers with system-level tasks.

Acknowledgment

This work was partially supported by AFOSR (Dynamics and Control, Drs. Jacobs and King) and the National Science Foundation.

Literature Cited

Åstrom, K. J., and B. Wittenmark, *Adaptive Control*, 2nd ed., Addison Wesley, New York (1995).

Cybenco, G., "Just in Time Learning and Estimation," *Identification, Adaptation, Learning*, S. Bittani and G. Picci, eds., NATO ASI, Springer, New York, pp. 423–434 (1996).

Gear, C. W., I. G. Kevrekidis, and C. Theodoropoulos, "Coarse Integration/Bifurcation Analysis via Microscopic Simulators," *Comp. Chem. Eng.*, **26**, 941 (2002).

Gillespie, D. T., "A General Method for Numerically Simulating the Stochastic Time Evolution of Coupled Chemical Reactions," *J. Comput. Phys.*, **22**, 403 (1976).

Gillespie, D. T., "Exact Stochastic Simulation of Coupled Chemical Reactions," *J. Phys. Chem.*, **81**, 2340 (1977).

Kailath, T., *Linear Systems*, Prentice Hall, Englewood Cliffs, NJ (1980).

Kalman, R. E., and R. S. Bucy, "New Results in Linear Filtering and Prediction Theory," *Trans. ASME, Ser. D, J. Basic Eng.*, **83**, 95 (1961).

Kevrekidis, I. G., C. W. Gear, J. M. Hyman, P. G. Kevrekidis, Runborg, O. and C. Theodoropoulos, "Equation-Free Multiscale Computation: Enabling Microscopic Simulators to Perform System-Level Tasks," *Comm. Math. Sciences*, in press (2003); (available on the Web at also http://www.arxiv.org/PS – cache/physics/pdf/0209/0209043.pdf)

Ljung, L., *System Identification: Theory for the User*, 2nd ed., Prentice Hall, NJ (1999).

Lou, Y., and P. D. Christofides, "Estimation and Control of Surface Roughness in Thin Film Growth Using Kinetic Monte-Carlo Models," *IEEE CDC*, Las Vegas, NV (2002).

Lust, K., "Numerical Bifurcation Analysis of Periodic Solutions of Partial Differential Equations," PhD thesis, Katholieke Universiteit Leuven (1997).

Makeev, A. G., D. Maroudas, and I. G. Kevrekidis, "Coarse Stability and Bifurcation Analysis using Stochastic Simulators: Kinetic Monte Carlo Examples," *J. Chem. Phys.*, **116**, 10083 (2002).

Rico-Martinez, R., K. Krischer, G. Flätgen, J. S. Anderson, and I. G. Kevrekidis, "Adaptive Detection of Bifurcations: An Experimental Feasibility Study," *Physica D*, **176**(1-2), 1–18 (2003).

Runborg, O., C. Theodoropoulos, and I. G. Kevrekidis, "Effective Bifurcation Analysis: a Time-Stepper-Based Approach," *Nonlinearity*, **15**, 491 (2002).

Sage, A. P., and C. C. White, *Optimum Systems Control*, 2nd ed., Prentice Hall, Englewood Cliffs, NJ (1977).

Savitzky, A., and M. J. E. Golay, "Smoothing and Differentiation of Data by Simplified Least Squares Procedures," *Anal. Chemistry*, **36**, 1627 (1964).

Shroff, G. M., and H. B. Keller, "Stabilization of Unstable Procedures—the Recursive Projection Method," *SIAM J. Num. Anal.*, **30**, 1099 (1993).

Siettos, C. I., A. Armaou, R. Rico-Martinez, A. G. Makeev, and I. G. Kevrekidis, "Some Issues at the Interface between Control and Microscopic/Stochastic Simulations," AIChE Meeting, Indianapolis, IN (2002).

Theodoropoulos, C., Y. H. Qian, and I. G. Kevrekidis, "Coarse Stability and Bifurcation Analysis using Time-Steppers: a Reaction-Diffusion Example," *PNAS*, **97**, 9840 (2000).

Tuckerman, L. S., and D. Barkley, "Bifurcation Analysis for Time-Steppers," *Numerical Methods for Bifurcation Problems and Large-Scale Dynamical Systems*, E. Doedel and L. S. Tuckerman, eds., Springer, New York (2000).

Manuscript received Aug. 30, 2002, and revision received Mar. 4, 2003.