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FINDING DOMINANT STRUCTURES OF NONREVERSIBLE MARKOV PROCESSES*

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Abstract. Finding metastable sets as dominant structures of Markov processes has been shown to be especially useful in modeling interesting slow dynamics of various real world complex processes. Furthermore, coarse graining of such processes based on their dominant structures leads to better understanding and dimension reduction of observed systems. However, in many cases, e.g. for nonreversible Markov processes, dominant structures are often not formed by metastable sets but by important cycles or mixture of both. This paper aims at understanding and identifying these different types of dominant structures for reversible as well as nonreversible ergodic Markov processes. Our algorithmic approach generalizes spectral based methods for reversible process by using Schur decomposition techniques which can tackle also nonreversible cases. We illustrate the mathematical construction of our new approach by numerical experiments.

Key words. nonreversible Markov processes, metastable sets, cycle decomposition, Schur decomposition

AMS subject classifications. 65C40, 60J20, 82C26

1. Introduction. Many complex processes in physics, chemistry, geo- and material sciences but also in the social sciences can be modelled by stochastic Markov processes. Mostly these processes exhibit on continuous and often high dimensional state spaces, and a rich hierarchy of temporal and spatial scales. Whenever one is not interested in full resolution of all scales, one aims at coarse graining the Markov process model in a way that still allows for an accurate description of the scales of interest while everything else is considered only collectively. In recent years it has been shown that under appropriate conditions coarse graining leads to *Markov models* on discrete (often even finite) state spaces. Well known examples are Markov State Models (MSMs) in molecular dynamics [23, 35, 34], kinetic Monte Carlo models in catalysis [38, 37] or biochemical reaction networks [36], or complex network modules in the social or biological sciences [18, 21, 29, 22].

Such Markov models can be used to find *dominant structures* of the underlying complex process. For example, Markov State Model (MSM) building in molecular dynamics (MD) allows for finding the *dominant metastable sets* of the underlying MD process [31, 34, 30] and through it a better understanding of biomolecular function, for reversible processes [7, 25, 35, 15, 16] as well as nonreversible ones [39]. In contrast, in catalytic processes the dominant structures are no longer formed by metastable sets but by important process cycles [19]; finding the *dominant cycles* of a kinetic Monte Carlo model allows for understanding the catalytic activity.

Stochastic processes are best understood in equilibrium where the process must necessarily be (1) time-homogeneous and (2) reversible. In other words, the evolution of the process must (1) not be driven by an external effect that changes with time and (2) be invariant or statistically indistinguishable under time reversal. MSM building,

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for one example, is understood to a high degree as long as the underlying process is in equilibrium; the spectral approaches to the algorithmic identification of metastable sets for reversible Markov chains is well-developed. For another example, module finding and clustering of complex networks are much better understood for undirected networks (where the associated random walk is time-homogeneous and reversible) than for directed networks where dominant cycles can take over the role of metastable sets. What is needed is an algorithmic approach that allows to identify dominant structures (metastable sets, dominant cycles, and mixtures of both) of Markov models for reversible as well as nonreversible processes.

The present article is presenting such an algorithmic approach. This approach is built to identify dominant structures of *nonequilibrium steady state* (NESS) Markov processes. The distinction between reversible and NESS processes is understood as follows: In a NESS process there still is a steady state, given by an invariant measure μ of the process wrt. which the process is ergodic. Provided the process is distributed according to μ at time $t = 0$, let $p_\tau(A, B)$ denote the probability to start at $t = 0$ from A and find the process in B at time $t = \tau$. Then reversibility means $p_\tau(A, B) = p_\tau(B, A)$, and for a NESS process this identity is violated in some cases such that there is an effective probability flow $p_\tau(A, B) - p_\tau(B, A) \neq 0$ between some regions (sets of states) A and B in state space.

Markov models of NESS processes still have the property that the probability flow must be divergent-free [20], i.e., for any region A , the total flow into A must equal the total flow out of A . Therefore the flow must be decomposable into elementary cycles [2]. Many authors have picked up the idea that these elementary cycles encode important information about NESS processes [14, 13]. However, an efficient algorithmic approach that seamlessly combines the cycle decomposition idea with identification of dominant structures (metastable sets and/or dominant cycles) is still missing. We will present such an approach for nonreversible ergodic processes; this will be achieved by answering two main questions:

- (Q1) What happens when we break the symmetry of a reversible process by perturbing it into a NESS process, or, more precisely, what kind of perturbation can turn a process in equilibrium with dominant metastable sets into a NESS process with dominant cycles (that perhaps coexist with metastable sets)?
- (Q2) The reversible case can be tackled with spectral approaches that are based on well-known and efficient numerical linear algebra schemes. Is there a way to use similarly efficient linear algebra numerics for tackling the nonreversible case, too?

The article is composed as follows: In Sec. 2 we shortly review the spectral approach to finding metastable sets for reversible Markov models. Next, in Sec. 3 we present the stochastic cycle decomposition of probability flows for Markov chains. Then, in Sec. 4, we discuss question (Q1) from above and show what kind of symmetry breaking is required to perturb a reversible chain into a NESS process with dominant cycles. Finally, in Sec. 5, we answer question (Q2) and present an algorithmic approach based on Schur decomposition techniques that allows for finding metastable sets as well as dominant cycles, and in Sec. 6 we discuss the performance of this algorithm in application to some numerical examples.

2. Spectral approach to finding metastable sets. In all of the following we consider an irreducible and aperiodic Markov chain of the finite state space $S = \{1, \dots, n\}$ that is given by the transition matrix P . In addition we assume that the associated invariant measure μ is positive everywhere. Then, μ is the unique

normalized ($\sum_i \mu_i = 1$) left eigenvector of P for eigenvalue $\lambda = 1$ while the right eigenvector simply is given by $e = (1, \dots, 1)^T \in \mathbb{R}^n$. Let D denote the diagonal matrix with $D_{ii} = \mu_i$, and let us define the weighted scalar product

$$\langle u, v \rangle = u^* D v.$$

The probability flow associated with the process is given by the flow matrix

$$F = DP,$$

i.e., the (steady state) probability flow from state i to j is $F_{ij} = \mu_i P_{ij}$. If F is not symmetric we have a NESS process since then there are state $i, j \in S$ so that $F_{ij} \neq F_{ji}$.

Reversible processes. If the process is reversible the detailed balance condition,

$$\mu_i P_{ij} = \mu_j P_{ji}, \quad \forall i, j \in S \quad (2.1)$$

is satisfied, i.e., it holds $DP = P^T D$. Then the flow matrix F is symmetric, $F = F^T$, i.e., the flows between every pair of states are balanced so that there is no effective flow. Moreover, the transition matrix P is symmetric wrt. the scalar product $\langle \cdot, \cdot \rangle$. As a consequence P is diagonalizable,

$$P = Q \Lambda Q^{-1} = Q \Lambda Q^T D,$$

with a diagonal matrix Λ containing the real-valued eigenvalues of P and a $\langle \cdot, \cdot \rangle$ -orthonormal matrix Q such that

$$Q^T D Q = \text{Id}.$$

Thus, the eigenvalues of P can be ordered $1 = \lambda_1 \geq \lambda_2 \geq \dots \geq \lambda_n \geq -1$ with eigenvectors $u_1 = e, u_2, \dots, u_n$ that form the columns of Q and thus are orthonormal wrt. $\langle \cdot, \cdot \rangle$.

Metastable sets and spectral approach. Here we will refer to approaches which are oriented towards a relation between spectral properties of the transition matrix P and a partitioning of a state space S into metastable sets of this process [34]. That is, we will consider $m < n$ sets, such that

$$M_i \cap M_j = \emptyset, \quad i \neq j \quad \text{and} \quad \cup_{i=1}^m M_i \subseteq S. \quad (2.2)$$

Sets M_1, \dots, M_m are called metastable sets, if the process remains for a long period of time inside of $M_i, i = 1, \dots, m$ before it exits quickly to another metastable set. The best possible partition of the state space in metastable sets has to maximize the joint metastability

$$\mathcal{D}(M_1, \dots, M_m) = \sum_{i=1}^m p(M_i, M_i),$$

where $p(M_i, M_i)$ is the residence probability in set M_i . For reversible processes, it is known [3, 4, 34, 33, 11] that if the decomposition into metastable sets M_1, \dots, M_m , $m < n$ exists, then the spectrum of P has m dominant eigenvalues $1 = \lambda_1 \geq \lambda_2 \geq \dots \geq \lambda_m$ and for the rest of the spectrum it follows [33, 34] that

$$\{\lambda_{m+1}, \dots, \lambda_n\} \subset B_r(0) \subset \mathbb{C}, \quad r < \lambda_m.$$

In turn, the existence of m dominant eigenvalues guarantees the existence of a metastable decomposition into m sets with high joint metastability. In this case, the eigenvectors $u_1 = e, u_2, \dots, u_m$ corresponding to the m dominant eigenvalues, contain information on the location of the metastable sets [3, 34, 11]. The following theorem establishes the relation between a given decomposition into metastable sets and spectral properties of the transition matrix P :

THEOREM 2.1 ([12]). *The joint metastability of an arbitrary full decomposition M_1, \dots, M_m of the state space is bounded from below and above by*

$$\lambda_1 + \delta_2^2 \lambda_2 + \dots + \delta_m^2 \lambda_m + c \leq \mathcal{D}(M_1, \dots, M_m) \leq \lambda_1 + \lambda_2 + \dots + \lambda_m, \quad (2.3)$$

where $c = \lambda_{m+1} (1 - \delta_2^2 + \dots + 1 - \delta_m^2)$ and δ_j is the error of the $\langle \cdot, \cdot \rangle$ -orthogonal projection of the eigenvector u_j onto the space spanned by the characteristic functions $D = \text{span}\{\mathbf{1}_{M_1}, \dots, \mathbf{1}_{M_m}\}$ of the sets,

$$\delta_j = \|Q^\perp u_j\|, \quad j = 2, \dots, m. \quad (2.4)$$

Following this and similar connections between metastable sets of reversible processes and the dominant spectrum of P , many different methods for finding dominant metastable sets have been developed [8, 32, 24, 29], while comparable approaches for NESS processes are almost totally missing with just a very few limited exceptions like [39, 1] and [34], Sec. 5.3. One of these methods for the reversible case, PCCA [8], directly takes the dominant eigenvectors as input and computes an almost optimal metastable decomposition; however this approach suffers from the problem that in many cases there are transition states that do not really belong to one of the metastable states but somehow support the transitions between them.

Eigenvector-based PCCA+. Instead of identifying $m < n$ metastable sets, in [9] it was proposed to linearly transform the m dominant eigenvectors $u_1 = e, u_2, \dots, u_m$ of the transition matrix P into a set of nonnegative, linearly independent membership vectors $\xi_1, \dots, \xi_m \in [0, 1]^n$. In [34], Thm. 13, the above Theorem 2.1 has been generalized to this case.

These membership vectors form the columns of a membership matrix $\chi \in [0, 1]^{n \times m}$. Each entry χ_{ij} denotes the degree of membership of state $i = 1, \dots, n$, with regard to the metastable cluster $j = 1, \dots, m$ (we use the phrase metastable cluster instead of metastable set in order to distinguish between the two concepts). The matrix χ is row stochastic. There are many possible linear transformations from the eigenvectors to a feasible set of membership vectors. The algorithm PCCA+ is a constrained optimization approach for finding the best feasible linear transformation [40, 28]. If \hat{Q} denotes the $n \times m$ -submatrix of Q consisting of the dominant eigenvectors of P , then $\chi = \hat{Q}\mathcal{A}$ is the linear relation between χ and \hat{Q} with the optimal, regular $m \times m$ -transformation matrix \mathcal{A} . A straight forward optimization criterion would be to maximize the metastability of the membership vectors, which is in the spirit of Theorem 2.1. With the linear combination idea the projection error in equation (2.4) vanishes because ξ_i and u_i span the same space, see also [40, 10]. According to [27], however, a further possible optimization criterion for \mathcal{A} is to aim at “crisp” membership vectors ξ_i . The membership vectors ξ_i should be as orthogonal as possible with regard to $\langle \cdot, \cdot \rangle$. In this case, the convex objective function that has to be maximized is the trace of a row-stochastic stiffness matrix $\mathcal{S} = \mathcal{S}(\chi)$,

$$\mathcal{S}_{ij}(\chi) = \frac{\langle \xi_i, \xi_j \rangle}{\langle \xi_i, \mathbf{1} \rangle}, \quad \xi_i = i\text{th row of } \chi$$

so that PCCA+ solves the constrained optimization problem

$$\max_{\mathcal{A} \in \mathbb{R}^{m \times m}} \text{trace}(\mathcal{S}(\hat{Q}\mathcal{A})), \quad \text{s.t. } \chi = \hat{Q}\mathcal{A} \geq 0 \text{ and } \sum_j \chi_{ij} = 1.$$

3. Cycle decomposition. In order to arrive at a cycle decompositions of the probability flow F we follow [14, 13] and [2, 1]:

First, we generalize the notion of edge flows to cycle flows, where we define cycles in the following way: A k -cycle γ on S is defined as an ordered sequence (up to cyclic permutations) of k connected states, $\gamma = (i_1, i_2, \dots, i_k)$ with $P_{i_j, i_{j+1}} > 0$ and $P_{i_k, i_1} > 0$ with length $|\gamma| = k$. Cycles that do not have self-intersections are called simple cycles. The set of all simple cycles is denoted by \mathcal{C} .

Second, we call a collection $\mathcal{C}_+ \subset \mathcal{C}$ of cycles γ with real positive weights $w(\gamma)$ a flow decomposition iff for every edge $(i, j) \in S^2$ we have

$$F_{ij} = \sum_{\gamma \supset (i, j)} w(\gamma), \quad (3.1)$$

where we write $\gamma \supset (i, j)$ if the edge (i, j) is in γ .

Third, and last, we consider the stochastic definition of weights introduced in [14, 13]. Let $(X_k)_{1 \leq k \leq T}$ be a realization of the Markov chain under consideration. We say that the process $(X_k)_{1 \leq k \leq T}$ passes through a cycle γ if it passes through all edges of a cycle γ in the correct order, but not necessarily consecutively meaning that excursions through one or more full new cycles are allowed. Let N_T^γ be the number of times $(X_k)_{1 \leq k \leq T}$ passes through a cycle γ up to time T . The limit

$$w(\gamma) := \lim_{T \rightarrow \infty} \frac{N_T^\gamma}{T} \quad (3.2)$$

exists almost surely [13] because of the assumed ergodicity, and allows to establish the unique cycle decomposition (\mathcal{C}_+, w) . We will refer to $w(\gamma)$ as an importance weight for the cycle γ . As a consequence of (3.1) we have

$$F = \sum_{\gamma \in \mathcal{C}_+} w(\gamma) C_\gamma, \quad (3.3)$$

where C_γ denotes the permutation matrix associated with the cycle.

If the process is reversible, F is symmetric because of the detailed balance condition. If not, then its anti-symmetric part

$$F^A = \frac{1}{2}(F - F^T)$$

does not vanish, i.e. $F^A \neq 0$. When we denote the reversed cycle of the cycle γ by $R\gamma$ then $C_{R\gamma} = C_\gamma^T$ and

$$F^A = \frac{1}{2} \sum_{\gamma \in \mathcal{C}} (w(\gamma) - w(R\gamma)) C_\gamma.$$

This implies that in the reversible case the cycle γ and the associated reversed cycle γ_R have the same weights, $w(\gamma) = w(\gamma_R)$, while this is not always the case for nonreversible processes.

Dominant cycles. In the most extreme case of nonreversibility the cycle decomposition is dominated by a single cycle. For example, take $n = 3$, cycle $\gamma = (1, 2, 3)$, and

$$P = \frac{1}{1+2\epsilon} \begin{pmatrix} \epsilon & 1 & \epsilon \\ \epsilon & \epsilon & 1 \\ 1 & \epsilon & \epsilon \end{pmatrix}.$$

Then, the invariant measure is constant so that $F = P = C_\gamma + \mathcal{O}(\epsilon)$. That is, the cycle γ is the only one with significant weight. We will call such a process *almost cyclic* and the cycle γ *dominant*.

The eigenvalues of the above 3×3 matrix are $\lambda_1 = 1$, $\lambda_2 = e^{i2\pi/3} + \mathcal{O}(\epsilon)$ and $\lambda_3 = e^{-i2\pi/3} + \mathcal{O}(\epsilon)$, i.e., they almost lie on the unit circle in the complex plane, close to the zeros of $z^3 = 1$.

4. Perturbing reversible Markov chains. Based on what we have seen until here we understand there is a deep relation between dominant metastable sets and real-valued eigenvalues close to $\lambda = 1$, as well as another deep relation between dominant cycles and complex-valued eigenvalues close to the unit circle in the complex plane. In this section we will try to understand how the situation of a reversible process with dominant metastable sets can be perturbed into a nonreversible, almost cyclic situation. To this end, we will investigate how the real-valued eigenvalues of a reversible process can be driven into the complex plane away from the real line.

Let P again denote the transition matrix of an ergodic reversible process with real-valued eigenvalues λ_j and eigenvectors u_j as introduced above and consider the perturbation

$$P_\epsilon = P + \epsilon L$$

of P with a generator matrix L that has non-negative off-diagonal entries and satisfies

$$\sum_j L_{ij} = 0,$$

for all states $i \in S$.

4.1. Perturbation of eigenvalues. Under the above conditions, the eigenvalues and eigenvectors of P_ϵ satisfy the asymptotic expansion

$$\begin{aligned} \lambda_j^\epsilon &= \lambda_j + \epsilon \eta_j + \mathcal{O}(\epsilon^2) \\ u_j^\epsilon &= \tilde{u}_j + \epsilon v_j + \mathcal{O}(\epsilon^2), \end{aligned} \tag{4.1}$$

where $\tilde{u}_j = u_j$ if λ_j is a simple eigenvalue. If λ_j is degenerate, say m -fold, then the associated eigenspace E_j is m -dimensional and $\tilde{u}_j^{(1)}, \dots, \tilde{u}_j^{(n)}$ form an orthonormal basis of E_j such that the spectral perturbation is smooth in ϵ .

Insertion into the eigenproblem and comparison of all terms in linear order in ϵ yields

$$P v_j + L \tilde{u}_j = \lambda_j v_j + \eta_j \tilde{u}_j. \tag{4.2}$$

Simple eigenvalue λ_j . In this case, multiplication of (4.2) from the left with u_j and utilization of the reversibility of P yield

$$\eta_j = \langle u_j, L u_j \rangle,$$

which shows that the perturbation results in a shift of the eigenvalue along the real line.

Degenerate eigenvalue λ_j . Let us assume that λ_j is two-fold (i.e. $\lambda_j = \lambda_{j+1}$), and that the two associated eigenvectors are u_j and u_{j+1} . Then

$$\tilde{u}_j^{(i)} = c_{i,1}u_j + c_{i,2}u_{j+1}$$

with appropriate coefficients $c_{i,j}$, $i, j = 1, 2$ such that $|c_{i,1}|^2 + |c_{i,2}|^2 = 1$ for $i = 1, 2$. This time, multiplication of (4.2) for $i = 1, 2$ from the left with u_j and u_{j+1} and utilization of the reversibility of P yields two sets of conditions,

$$\begin{aligned} c_{1,1}\langle u_j, Lu_j \rangle + c_{1,2}\langle u_j, Lu_{j+1} \rangle &= \eta_j c_{1,1} \\ c_{1,1}\langle u_{j+1}, Lu_j \rangle + c_{1,2}\langle u_{j+1}, Lu_{j+1} \rangle &= \eta_j c_{1,2} \\ c_{2,1}\langle u_j, Lu_j \rangle + c_{2,2}\langle u_j, Lu_{j+1} \rangle &= \eta_j c_{2,1} \\ c_{2,1}\langle u_{j+1}, Lu_j \rangle + c_{2,2}\langle u_{j+1}, Lu_{j+1} \rangle &= \eta_j c_{2,2}, \end{aligned}$$

with suitable η_j . As a consequence the first order perturbations η_j are given by the eigenvalues of the matrix

$$\hat{L} = \begin{pmatrix} \langle u_j, Lu_j \rangle & \langle u_j, Lu_{j+1} \rangle \\ \langle u_{j+1}, Lu_j \rangle & \langle u_{j+1}, Lu_{j+1} \rangle \end{pmatrix}.$$

That is, the two eigenvalues η_j^+ and η_j^- of \hat{L} control the perturbation of the two-fold eigenvalue λ_j : If they are identical the eigenvalue remains two-fold (to first order). If $\eta_j^+ \neq \eta_j^-$ the eigenvalue is split into two simple eigenvalues which satisfies to first order

$$\lambda_j^{\epsilon, \pm} = \lambda_j + \epsilon \eta_j^{\pm}. \quad (4.3)$$

Complex or real eigenvalues? The two eigenvalues $\eta_j^+ \neq \eta_j^-$ are either both real-valued or come as a complex conjugated pair, and thus the perturbation either leads to a split along the real line or into the complex plane away from the real line. We have

$$\eta_j^{\pm} = \frac{1}{2}(\hat{L}_{11} + \hat{L}_{22}) \pm \left(\frac{1}{4}(\hat{L}_{11} - \hat{L}_{22})^2 + \hat{L}_{12}\hat{L}_{21} \right)^{1/2}.$$

Thus, the perturbation drives the two-fold real eigenvalue into the complex plane iff

$$\frac{1}{4}(\hat{L}_{11} - \hat{L}_{22})^2 < -\hat{L}_{12}\hat{L}_{21}. \quad (4.4)$$

Now let us consider the anti-symmetric part $\hat{L}^A = (\hat{L} - \hat{L}^T)/2$ of \hat{L} ,

$$\hat{L}^A = \frac{1}{2} \begin{pmatrix} 0 & \delta \\ -\delta & 0 \end{pmatrix},$$

with $\delta = \hat{L}_{12} - \hat{L}_{21}$ that measures the degree of deviation from symmetry of \hat{L} . In terms of this asymmetry, condition (4.4) can be rewritten as

$$\delta^2 = (\hat{L}_{12} - \hat{L}_{21})^2 > \frac{1}{2}(\hat{L}_{11} - \hat{L}_{22})^2 + \hat{L}_{12}^2 + \hat{L}_{21}^2. \quad (4.5)$$

Thus, the perturbation drives the two-fold real eigenvalue into the complex plane iff the deviation from symmetry of \hat{L} is strong enough.

Based on conditions (4.4) or (4.5) one can now check which kind of perturbation matrix L will drive a double eigenvector of the unperturbed, reversible transition matrix P into the complex plane. In the appendix we show that perturbations of a single entry (1-cycle) of P or of a 2-cycle can never lead to this kind of perturbation.

4.2. Illustrating example. Let us consider the reversible transition matrix

$$P = \begin{pmatrix} 0.8566 & 0.1195 & 0.0239 \\ 0.0566 & 0.9195 & 0.0239 \\ 0.0566 & 0.1195 & 0.8239 \end{pmatrix},$$

with eigenvalues $1 = \lambda_1$ and $\lambda_2 = \lambda_3 = 0.8$.

First, we study what happens if we perturb P using the 2-cycle $\gamma = (1, 2)$, that is we set

$$L_1 = \sigma_1 \begin{pmatrix} -1 & 1 & 0 \\ 1 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix} = \sigma(C_\gamma - I_\gamma), \quad \sigma_1 = 0.1242,$$

where I_γ denotes the diagonal matrix with 1 on the diagonal for all states that are involved in the cycle γ and zeros on all other diagonal entries.

Then, $\hat{P}_\epsilon = P + \epsilon L_1$ is still diagonalizable with real-valued eigenvalues $1 = \lambda_1$, $\lambda_2 = 0.8$, and

$$\lambda_3 \begin{cases} < 0.8 & \text{if } \epsilon > 0 \\ = 0.8 & \text{if } \epsilon = 0 \\ > 0.8 & \text{if } \epsilon < 0 \end{cases}$$

as long as \hat{P}_ϵ is stochastic.

Next, we perturb P along the 3-cycle $\Gamma = (1, 2, 3)$,

$$L_2 = \sigma_2(C_\Gamma - I_\Gamma), \quad \sigma_2 = 0.4119,$$

then $P_\epsilon = P + \epsilon L_2$ has the spectrum shown in Fig. 4.1 which clearly shows that starting from $\epsilon = 0$ (for which $\hat{P}_\epsilon = P$ has a double eigenvalue 0.8) the second and third eigenvalues develop imaginary parts and are driven into the complex plane for $\epsilon > 0$. In this case the first order perturbation result (4.3) for the two-fold eigenvalue $\lambda = 0.8$ can be checked by computing \hat{L} and its eigenvalues η^\pm . In this case, the result $0.8 + \epsilon\eta^\pm$ is graphically indistinguishable from the second and third eigenvalues of P_ϵ in $\epsilon \in [0, 1]$.

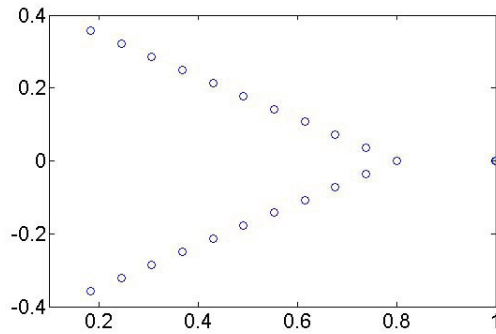


FIGURE 4.1. Spectrum of $P_\epsilon = P + \epsilon L_2$ for $\epsilon \in [0, 1]$.

4.3. Perturbation of probability flow. The perturbed flow matrix is given by $F_\epsilon = D_\epsilon P_\epsilon$, where D_ϵ denotes the diagonal matrix with entries given by the invariant measure μ_ϵ of P_ϵ .

The stochastic cycle decomposition of F^ϵ reads

$$F^\epsilon = \sum_{\gamma} w^\epsilon(\gamma) C_\gamma.$$

Its antisymmetric part F_A^ϵ satisfies

$$F_A^\epsilon = \epsilon(NP - P^T N + DL - L^T D) + \mathcal{O}(\epsilon^2),$$

if we use the asymptotic expansion of D_ϵ , $D_\epsilon = D + \epsilon N + \mathcal{O}(\epsilon^2)$.

With \hat{Q} denoting the $n \times 2$ -matrix that has the two eigenvectors u_j and u_{j+1} associated with the double eigenvalue of P as its columns we have

$$\hat{L} = \hat{Q}^T DL \hat{Q},$$

and (up to first order in ϵ)

$$A^\epsilon = \hat{Q}^T F_A^\epsilon \hat{Q}^T = \frac{\epsilon}{2}(\hat{L} - \hat{L}^T) = \epsilon \hat{L}^A = \frac{\epsilon}{2} \begin{pmatrix} 0 & \delta \\ -\delta & 0 \end{pmatrix}. \quad (4.6)$$

This can be seen from

$$\begin{aligned} A^\epsilon &= \frac{\epsilon}{2} \left(\hat{Q}^T (NP - P^T N) \hat{Q} + \hat{Q}^T (DL - L^T D) \hat{Q} \right) + \mathcal{O}(\epsilon^2) \\ &= \frac{\epsilon}{2} \left(\hat{Q}^T (NP - P^T N) \hat{Q} + 2\hat{L}^A \right) + \mathcal{O}(\epsilon^2), \end{aligned}$$

where we can use $P\hat{Q} = \lambda\hat{Q}$ and $\hat{Q}^T P^T = \lambda\hat{Q}^T$ to show that the term containing $NP - P^T N$ vanishes.

Interpretation. Together with (4.5) equation (4.6) shows, that the perturbation drives the two-fold real eigenvalue into the complex plane iff the deviation from symmetry of the perturbed flow matrix is strong enough.

4.4. Perturbation of the Schur decomposition. The real-valued Schur decomposition is an alternative to the spectral decomposition if P_ϵ is no longer reversible. Let P again be reversible with a double eigenvalue λ while the rest of the eigenvalues are simple. Then, the Schur decomposition (X_ϵ, R_ϵ) of the transition matrix $P_\epsilon = P + \epsilon L$ has the form

$$P_\epsilon X_\epsilon = X_\epsilon R_\epsilon, \quad \text{s.t.} \quad X_\epsilon^T D_\epsilon X_\epsilon = \text{Id}, \quad (4.7)$$

so that the columns of X_ϵ form an $\langle \cdot, \cdot \rangle$ -orthonormal basis (as the eigenvectors did for reversible P_ϵ) and where R_ϵ has the following form

$$R_\epsilon = \begin{pmatrix} B_\epsilon & U_\epsilon \\ 0 & \tilde{\Lambda}_\epsilon \end{pmatrix}. \quad (4.8)$$

B_ϵ denotes the 2×2 -block that belongs to the two-dimensional subspace that originates from the two-dimensional eigenspace of $\lambda = \lambda_j$ at $\epsilon = 0$, and $\tilde{\Lambda}_\epsilon$ is the diagonal matrix that contains the rest of the eigenvalues of P_ϵ (where we assume that ϵ is kept small enough such that only λ is perturbed into the complex plane and all other eigenvalues

stay real-valued and simple), and U_ϵ denotes all the remaining entries in the upper triangular part of R_ϵ (but not the ones contained in B_ϵ).

For $\epsilon = 0$ the Schur decomposition of $P_\epsilon = P$ simply is identical to the spectral decomposition discussed above.

In general, the eigenvalues of P_ϵ and R_ϵ are identical. Therefore the upper block, B_ϵ , of R_ϵ will encode whether the perturbation L forces a pair of eigenvalues into the complex plane. This is indeed true as the following results shows.

THEOREM 4.1. *Up to first order in ϵ the upper 2×2 block of the Schur form R_ϵ of P_ϵ is given by $\lambda \text{Id}_{2 \times 2} + \epsilon \hat{L}_s$ where \hat{L}_s denotes the Schur form of \hat{L} (and thus has the same eigenvalues). Thus, the Schur form \hat{L}_s of the projection of L to the eigenspace of a double eigenvalue of the reversible matrix P is sufficient to see whether the perturbation leads to a complex conjugated pair of eigenvalues.*

Proof. First we consider the asymptotic expansion of R_ϵ that is given by

$$\begin{aligned} R_\epsilon &= \Lambda + \epsilon \mathcal{R} + \mathcal{O}(\epsilon^2) \\ X_\epsilon &= X_0 + \epsilon Y + \mathcal{O}(\epsilon^2), \end{aligned}$$

where Λ is diagonal with λ on the first two diagonal entries and then the rest of the eigenvalues of P in the order of $\tilde{\Lambda}_0$.

If we put the asymptotic expansion into $P_\epsilon X_\epsilon = X_\epsilon R_\epsilon$ we get in first order of ϵ that

$$PY + LX_0 = Y\Lambda + X_0\mathcal{R}, \quad (4.9)$$

Since the columns of X_0 form a basis, there is a coefficient matrix A such that

$$Y = X_0 A.$$

Inserting this into (4.9) and using $PX_0 = X_0\Lambda$ yields

$$X_0\Lambda A + LX_0 = X_0\Lambda A + X_0\mathcal{R},$$

from which by multiplication with $X_0^T D$ from the left we get

$$\Lambda A + L_s = \Lambda A + \mathcal{R}, \quad \text{with } L_s = X_0^T D L X_0.$$

Written in terms of the entries A_{kl} of A this reads

$$A_{kl}(\Lambda_{kk} - \Lambda_{ll}) = \mathcal{R}_{kl} - L_{s,kl}. \quad (4.10)$$

First we consider $k, l \in \{1, 2\}$. Then $\Lambda_{kk} = \Lambda_{ll} = \lambda$ and thus we get

$$\mathcal{R}_{kl} = L_{s,kl}, \quad k, l \in \{1, 2\}.$$

Next, we denote the first two columns of X_0 by \hat{X}_0 such that the upper 2×2 block of L_s can be written

$$\hat{L}_s = \hat{X}_0^T D L \hat{X}_0,$$

and we get that the upper 2×2 block B_ϵ of R_ϵ has the asymptotic form

$$B_\epsilon = \lambda \text{Id}_{2 \times 2} + \epsilon \hat{L}_s + \mathcal{O}(\epsilon^2). \quad (4.11)$$

\hat{X}_0 spans the same subspace as \hat{Q} so that \hat{L}_s is the Schur form of \hat{L} (and thus has the same eigenvalues).

Now we consider the indices $k = l > 2$ in (4.10) we get $\mathcal{R}_{kk} = L_{s,kk}$ and thus

$$\tilde{\Lambda}_{\epsilon,kk} = \lambda_k + \epsilon u_k^T D L u_k + \mathcal{O}(\epsilon^2), \quad (4.12)$$

where u_k denotes the k th column, $k = 3, 4, \dots$, of X_0 which is identical to the eigenvector of P associated with the eigenvalue λ_k .

Result. The Schur decomposition offers an alternative to the spectral decomposition since the Schur basis X_ϵ is identical to the eigenvector basis for all simple eigenvalues while the two basis-vectors related to the 2×2 diagonal block form a real-valued basis of the two-dimensional eigenspace that is associated with the complex-conjugated pair of eigenvalues.

5. Schur decomposition and dominant structures. Our aim: The large state space S with $|S| = n \gg 1$ states has to be decomposed into a smaller number $m \ll n$ of clusters of states that belong to dominant structures of the chain. One well-known example is the partitioning of S into metastable sets or metastable clusters described in Section 2 above. The idea of this article is to cluster states of S not (only) according to their metastability, but to cluster them according to a common transition pattern. We will re-use the idea of PCCA+, i.e., the decomposition of S is done by using membership vectors $\xi_1, \dots, \xi_m \in [0, 1]^n$ instead of sets. However, instead of just aiming at metastable clusters we will be aiming at identifying the dominant cycles as well. In analogy to the case of eigenvector-based PCCA+ for reversible processes, dominant structures will be defined by utilizing the dominant Schur vectors $\hat{X} \in \mathbb{R}^{n \times m}$ of the transition matrix P instead of the eigenvectors. According to the PCCA+ idea, we will aim at a linear combination of these vectors and define a membership matrix $\chi = \hat{X}\mathcal{A} \in [0, 1]^{n \times m}$. As in eigenvector-based PCCA+, the optimization criterion is to maximize the crispness of χ . We will show in the following that this type of objective function preserves the nonreversibility of the flow.

Projected Markov process. The real Schur decomposition of a general nonreversible ergodic transition matrix P has the form $PX = XR$ with $X^TDX = \text{Id}$. If we take an $m \times m$ -submatrix $\hat{R} \in \mathbb{R}^{m \times m}$ of R consisting of the top left part of R and not splitting a 2×2 -block of R , then

$$P\hat{X} = \hat{X}\hat{R}$$

and $\hat{X}^TD\hat{X} = \text{Id}$ holds, where \hat{X} consists of the first m columns of X . These vectors will be denoted as the dominant Schur vectors \hat{X} . The PCCA+ idea is to find a matrix \mathcal{A} such that the membership vectors are given by the columns of $\chi = \hat{X}\mathcal{A}$. According to [17, 34], the projected transition matrix \hat{P} is then defined as

$$\hat{P} = (\chi^TD\chi)^{-1}(\chi^TD P \chi) = \mathcal{A}^{-1}\hat{R}\mathcal{A}.$$

By this equation, it holds that

$$\hat{P}\mathcal{A}^{-1} = \mathcal{A}^{-1}\hat{R}.$$

Thus, \hat{P} has a Schur decomposition with the real Schur matrix \hat{R} . The corresponding Schur vectors stem from an orthonormalization of the column vectors of \mathcal{A}^{-1} . In this way, the dominant part \hat{R} of the Schur matrix R of P becomes the Schur matrix of the projected transition matrix \hat{P} , see also [43].

Sorting the Schur values. A real Schur decomposition is not unique. The reason is, that the diagonal elements (and 2×2 -blocks) of the Schur matrix R can be sorted in different ways, leading to different Schur vectors X . How to sort these values? When interested in finding dominant structures then the projected transition matrix \hat{P} should be close to a $\{0, 1\}$ -transition matrix, i.e., up to small perturbations the process has a dominant (almost deterministic) flow. Finding dominant cycles and metastabilities, therefore, implies that \hat{P} should be close to a permutation matrix. Permutation matrices, however, have eigenvalues on the unit circle. Since the Schur

values of the projected matrix coincide with the Schur values of the selected invariant subspace of P , the Schur values of P should be sorted according to their distance to the unit circle. They should be sorted with decreasing absolute value of the corresponding eigenvalues of P . Brandts [6] has formulated a software code for sorting the Schur decomposition accordingly.

Dominant structures and cycles. In this section, we are interested in the non-reversibility of the projected flow. There are in principle two possibilities. First, the projection might hide the non-reversibility of the original flow, because it only regards the flow between the clusters not within the clusters. Second, the projection can inherit the non-reversibility of the flow by leading to a transition matrix \hat{P} which is close to a (non-unit) permutation matrix. If one analyzes \hat{P} as an “independent” transition matrix, the previous sections have shown that the non-reversibility of a transition matrix \hat{P} is connected to the non-symmetry of the diagonal blocks of its Schur matrix \hat{R} . The low-dimensional matrix \hat{P} , however, stems from a clustering on a large state space. The question remains whether the non-symmetry of \hat{R} is also connected to the non-reversibility of the projected flow. We will see that aiming at such a direct relation, implies to use the “crispness” objective function by Roebnitz for the PCCA+ algorithm.

Non-reversibility of the projected flow. The flow F is projected to the clusters $\chi = \hat{X}\mathcal{A}$ of the PCCA+ algorithm. The non-reversibility of the projected flow is expressed by the matrix $\chi^T(F - F^T)\chi$. A weighted Frobenius norm $r = \|\hat{D}^{-1/2}\chi^T(F - F^T)\chi\hat{D}^{-1/2}\|_F$ of this matrix is used in order to quantify the non-reversibility r . In this norm, $\hat{D} = \text{diag}(\chi^T\mu)$ is the diagonal matrix of the projected invariant measure μ of P . With these preparations, the following equations hold

$$\begin{aligned} r &= \|\hat{D}^{-1/2}\chi^T(F - F^T)\chi\hat{D}^{-1/2}\|_F \\ &= \|\hat{D}^{-1/2}\mathcal{A}^T(\hat{X}^T F \hat{X} - \hat{X}^T F^T \hat{X})\mathcal{A}\hat{D}^{-1/2}\|_F \\ &= \|\hat{D}^{-1/2}\mathcal{A}^T(\hat{R} - \hat{R}^T)\mathcal{A}\hat{D}^{-1/2}\|_F \\ &= \sqrt{2 \sum_{i < j} |\hat{R}_{ij} - \hat{R}_{ji}|^2 \cdot \|M^{(i)} \times M^{(j)}\|_2^2}, \end{aligned} \quad (5.1)$$

where $M^{(i)}$ denotes the i -th row of $M = \mathcal{A}\hat{D}^{-1/2}$. In (5.1), each element-wise non-symmetry $|\hat{R}_{ij} - \hat{R}_{ji}|^2$ is weighted with the squared Euclidean norm of a cross product $\|M^{(i)} \times M^{(j)}\|_2^2$. By this term, the quality of the clustering enters this equation. What is the size of these weights? As explained in Section 2, PCCA+ can be used to maximize the orthogonality of the membership functions ξ_i with regard to the weighted inner product $\langle \cdot, \cdot \rangle$. The row stochastic mass matrix $\mathcal{S} = \hat{D}^{-1}\chi^T D \chi = \hat{D}^{-1}\mathcal{A}^T \mathcal{A}$ should be as close as possible to a unit matrix. If \mathcal{S} would be a unit matrix $\mathcal{S} = \text{Id}$, then $\text{Id} = \hat{D}^{-1}\mathcal{A}^T \mathcal{A}$ and, thus, $\text{Id} = \hat{D}^{-1/2}\mathcal{A}^T \mathcal{A}\hat{D}^{-1/2}$. In this case, the matrix $\hat{D}^{-1/2}\mathcal{A}^T$ would be orthogonal, which means that the rows of M would form an orthonormal basis. All the cross products would be identical $\|M^{(i)} \times M^{(j)}\|_2^2 = 1$. Thus, if we take the crispness function as the optimization criterion for PCCA+, we aim at the following: The non-reversibility of the flow should equal the non-symmetry of the reduced Schur matrix \hat{R} . In that sense, taking Schur vectors for PCCA+ preserves the non-reversibility of the flow.

6. Numerical Examples.

6.1. From metastable sets to dominant cycles in a nutshell. We consider the rate matrix

$$R = \begin{pmatrix} -2.0010 & 0.2773 & 0.7667 & 0.9432 & 0.0138 \\ 0.1309 & -2.9073 & 0.7523 & 1.5372 & 0.4868 \\ 0.0162 & 0.0336 & -0.0740 & 0.0026 & 0.0217 \\ 0.0281 & 0.0970 & 0.0036 & -0.1399 & 0.0111 \\ 0.0004 & 0.0328 & 0.0328 & 0.0118 & -0.0779 \end{pmatrix},$$

which has a double eigenvalue $\hat{\lambda} = -0.1$:

$$\lambda = 0, -0.1, -0.1, -2, -3.$$

Next we define $P = \exp(tR)$ with $t = 1$ which leads to

$$P = \begin{pmatrix} 0.1437 & 0.0401 & 0.3494 & 0.4344 & 0.0323 \\ 0.0189 & 0.0740 & 0.2585 & 0.4861 & 0.1625 \\ 0.0074 & 0.0115 & 0.9385 & 0.0184 & 0.0242 \\ 0.0130 & 0.0307 & 0.0261 & 0.9094 & 0.0209 \\ 0.0010 & 0.0109 & 0.0365 & 0.0223 & 0.9292 \end{pmatrix},$$

with eigenvalues

$$\lambda = 1, 0.9048, 0.9048, 0.1353, 0.0498,$$

where $\lambda = \exp(\hat{\lambda}) = 0.9048$ also is a double eigenvalue.

In order to perturb the double eigenvalue λ into the complex plane we consider the single cycle perturbation

$$L = \sigma(C_\gamma - I_\gamma), \quad \gamma = (3, 4, 5), \quad \sigma = 0.9,$$

and set

$$P_\epsilon = P + \epsilon L.$$

One finds

$$\hat{L} = \begin{pmatrix} -1.270 & 0.724 \\ -0.833 & -1.497 \end{pmatrix},$$

such that $-\hat{L}_{12}\hat{L}_{21} = 0.604$ and $(\hat{L}_{11} - \hat{L}_{22})^2/4 = 0.005$ so that the perturbation will lead into the complex plane. Fig. 6.1 shows the eigenvalues of P_ϵ in their dependence on ϵ . We see that the perturbation is well approximated by the first order result (4.3).

We observe that for $\epsilon = 0$ the process is reversible with three metastable sets (and three real-valued eigenvalues close to $\lambda = 1$), while for $\epsilon = 1$ it shows strongly irreversible, cyclic behavior with dominant cycle (3, 4, 5). Due to this the kinetics of the process changes: If we start with a probability distribution $u_0 = (0, 0, 1, 0, 0)$ that locates the process solely in state 3 at $k = 0$, then the evolution of this initial distribution under the process,

$$u^\epsilon(k) = P_\epsilon^k u_0,$$

leads to an evolution of the probability u_3^ϵ to be in state 3 as shown in Fig. 6.2. For $\epsilon = 0$ we observe the slow exponential decay that we expect in the presence of

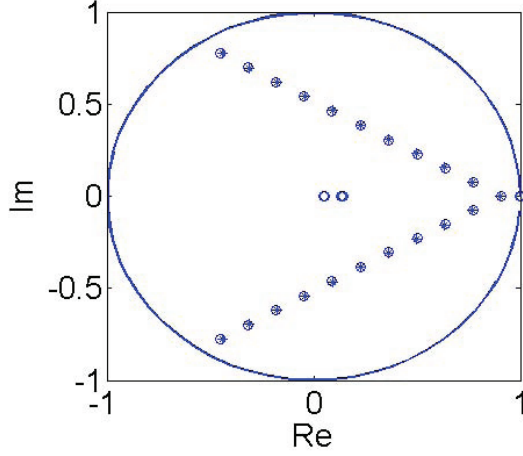


FIGURE 6.1. Spectrum of P_ϵ for $\epsilon \in [0, 1]$ in the unit circle of the complex plane (real- versus imaginary values). Circles: Eigenvalues of P_ϵ . Stars: First order perturbation of eigenvalues of P_ϵ .

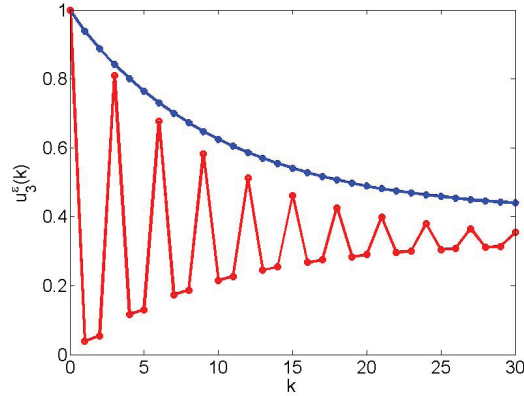


FIGURE 6.2. Evolution of the probability to be in state 3 under $P_0 = P$ (u_3^0 ; blue stars) and $P_1 = P + L$ (u_3^1 ; red circles) versus the step number k .

metastable sets for a reversible process, while for $\epsilon = 1$ we see the slowly damped oscillatory behavior that is characteristic for dominant cycles. In both cases the invariant measure of State 3 is reached for $k \rightarrow \infty$.

The change of dominant structures from three metastable sets (for $\epsilon = 0$) to a dominant cycle $(3, 4, 5)$ (for $\epsilon = 1$) can be observed by following the changing cycle weights (3.2) for increasing ϵ , under $P_\epsilon = P + \epsilon L$, as shown in Figure 6.3. For the reversible process, i.e., transition matrix $P = P_0$, metastability of sets $M_1 = \{3\}$, $M_2 = \{4\}$ and $M_3 = \{5\}$ (which are 1-cycles) can be seen from the high entries on the diagonal of P . As ϵ is increasing, the weights of these sets decrease towards 0 as their metastability decreases. On the other hand, the weight of a cycle $\gamma = (3, 4, 5)$ increases, and γ is becoming a dominant cycle for $\epsilon = 1$. The perturbation L does not influence other structures in our system, leaving the weights of all other cycles

almost constant for different values of ϵ .

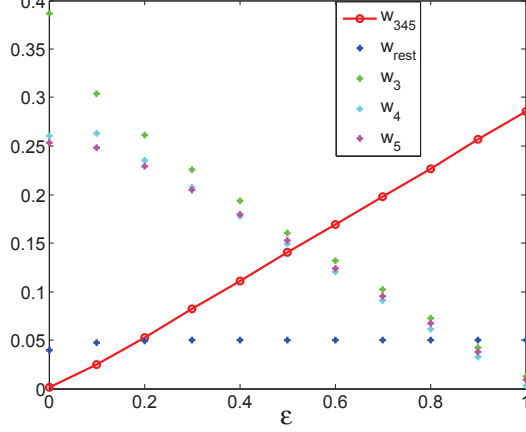


FIGURE 6.3. Evolution of cycle weights under $P_\epsilon = P + \epsilon L$: weight $w(\gamma)$ of the cycle $(3, 4, 5)$ in red, weight of a 1-cycle (3) in green, weight of a 1-cycle (4) in cyan, weight of a 1-cycle (5) in pink, sum of weights of all other cycles in blue.

Consider now the projection \hat{P} of the transition matrices P by Schur-based PCCA+ with $m = 3$ clusters. In this case we will sort the Schur matrix R according to Section 5, i.e., according to the absolute value of the corresponding eigenvalues. The objective function for an optimal linear transform from the corresponding leading three Schur vectors \hat{X} to the 5×3 -membership matrix χ is given by the crispness criterion. The reduced 3×3 -Schur matrix \hat{R} comprises of (at most) one 2×2 -block with $\hat{R}_{32}, \hat{R}_{23} \neq 0$ quantifying the non-symmetry of \hat{R} . As explained in Section 5, this non-symmetry $|\hat{R}_{23} - \hat{R}_{32}|^2$ quantifies also the non-reversibility of the projected flow. In order to measure the relation between the non-symmetry of \hat{R} and the non-reversibility of the projected flow, the term $|\hat{R}_{23} - \hat{R}_{32}|^2$ has to be weighted with a cross product $\|M^{(2)} \times M^{(3)}\|_2^2$. This weight and the optimal value of the objective function of PCCA+ are shown in Fig. 6.4. One clearly can see that the weight correlates well with the optimal value of the objective function, which can be 1 at maximum.

For extreme values $\epsilon \approx 0$ or $\epsilon \approx 1$, the clustering of PCCA+ nearly takes the maximally possible value, i.e., the three clusters of the system are well-separated from each other. In all cases the clustering identifies the states $\{3, 4, 5\}$ to be the centers of the clusters and $\{1, 2\}$ to belong to these clusters with a varying degree of membership. For $\epsilon \approx 0$, the clusters are metastable clusters of P_ϵ . For $\epsilon \approx 1$, the clusters form a dominant cycle of P_ϵ . For in between ϵ -values the matrix \hat{P}_ϵ is a mixture between a unit matrix (metastability) and a permutation matrix (cycle). For the extreme ϵ values the matrix \hat{P}_ϵ is close to a $\{0, 1\}$ -stochastic matrix, such that their rows have a small entropy, see also Fig. 6.5. Schur-based PCCA+ preserves the non-reversibility (dominant cyclic structure) of the flow.

6.2. Langevin Dynamics. For this illustrative example we perform a low-friction Langevin dynamics simulation of a double-well potential [26]:

$$\dot{r} = p, \quad \dot{p} = -\nabla_r V(r) - \gamma p + \sigma \dot{W}_t, \quad (6.1)$$

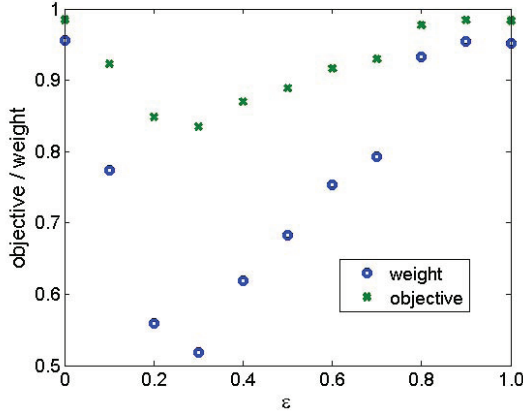


FIGURE 6.4. The optimal value of the PCCA+ objective function $\text{trace}(S)$ is plotted against ϵ (crosses). The weight $\|M^{(2)} \times M^{(3)}\|_2^2$ is plotted using circles and correlates well with the objective.

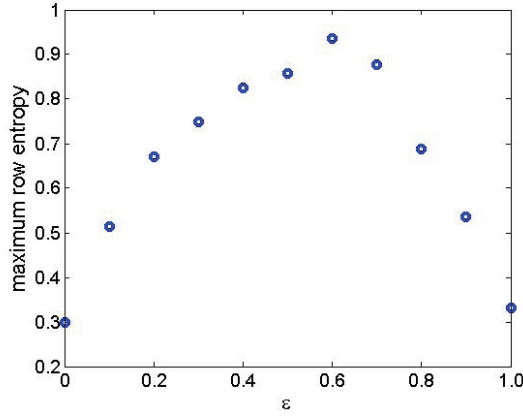


FIGURE 6.5. The maximum entropy of the rows of the projected transition matrix \hat{P}_ϵ of P_ϵ as a function of ϵ . Zero entropy corresponds to a $\{0,1\}$ -stochastic matrix, i.e., to a deterministic process. Thus, the matrix \hat{P}_ϵ is close to the unit or to a permutation matrix in our example for the low-entropy case (extreme ϵ -values). High entropy values indicate a mixture between a metastable and an almost cyclic system (in between ϵ -values).

where r is the vector of the Euclidean coordinates of all atoms in the molecular system, p the vector of associated momenta, $V(r) = (r^2 - 1)^2$ the interaction energy and $-\nabla_r V(r)$ the vector of all inter-atom interaction forces. $F_{\text{ext}} = \sigma \dot{W}_t$ is the external forcing given by a $3N$ -dimensional Brownian motion W_t and we set the friction constant $\gamma = 0.2$. The Langevin process (Y_t) is not reversible and the typical transition from the vicinity of one of the wells across the energy barrier at $r = 0$ towards the other well will look as follows: First the trajectory will orbit the initial well for some period of time before it crosses the barrier and starts to orbit the target well until it finally hits the close vicinity around the respective energy minimum.

The two-dimensional state space $[-1.8, 1.8] \times [-1.8, 1.8]$ has been discretized into 342 boxes B_i of size $\Delta r = 0.2$ and $\Delta p = 0.2$. The transition probabilities have been

computed using $M = 100$ simulated trajectories of length $\tau = 0.25$ [5, 34]. Note that $\tau = 0.25$ is very small compared to the expected transition time (which is larger than 100 here) and still shorter than the period of the periodic orbits of the system. The result is a 342×342 -transition matrix

$$P_{ij} = \frac{m_{ij}}{M},$$

where m_{ij} is the number of trajectories starting in B_i and ending up in B_j .

The invariant distribution $D = \text{diag}(\mu)$ of this matrix is used for the computation of the Schur decomposition sorted according to the absolute value of the corresponding eigenvalues. In order to include all real eigenvalues of P close to $\lambda_1 = 1$, we have to take $m = 9$. Thus, we used PCCA+ to compute the membership matrix χ based on the 342×9 -matrix \hat{X} of dominant Schur vectors and determined the projected transition matrix \hat{P} between the clusters.

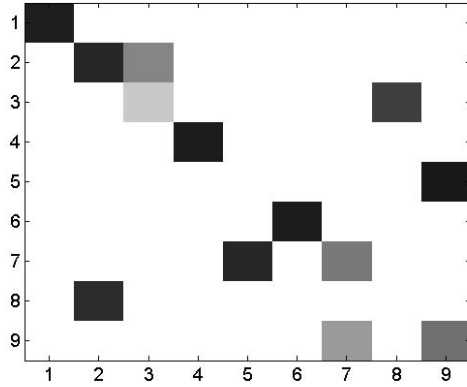


FIGURE 6.6. The 9-by-9-transition matrix \hat{P} between the clusters of the Langevin process. \hat{P} is not diagonally dominant but displays metastable as well as cyclic structures.

The resulting \hat{P} is shown in Fig. 6.6 and has the following interpretation. Clusters No. 1, 4, and 6 are metastable clusters. In order to visualize these clusters, we plot the $18 \times 19 = 342$ discretization boxes using a color according to the entries of the columns (1,4, and 6) of the membership matrix χ , see Fig. 6.7.

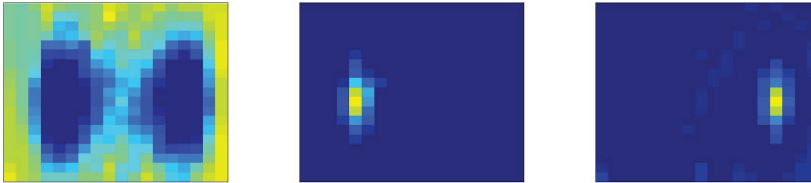


FIGURE 6.7. Membership values according to cluster no. 1 (left), 4 (middle) and 6 (right). These clusters are metastable clusters of the system. Blue color means low membership value and yellow high membership value. Cluster no. 1 is the “rapidly-mixing” high-energy cluster. The clusters no. 4 and no.6 are the low-energy clusters at the basins of the double-well potential and at low absolute momentum value.

The other clusters belong to dominant cycles. The cycles can be extracted from the matrix \hat{P} shown in Fig. 6.6. One cycle is $2 \rightarrow 3 \rightarrow 8 \rightarrow 2$, the other cycle is $5 \rightarrow 9 \rightarrow 7 \rightarrow 5$. In Fig. 6.8, one of these cycles is visualized. The other cycle is symmetric to one shown.

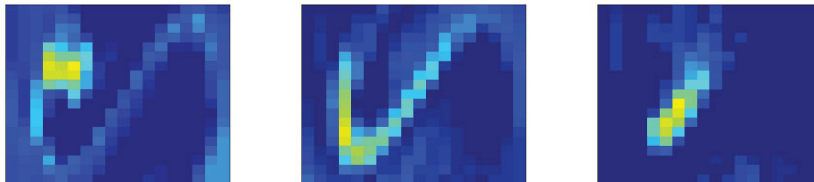


FIGURE 6.8. From left to right: Visualization of the clusters no. 2, 3, and 8. These clusters represent a dominant cycle of the Langevin dynamics inside the left basin of the double-well potential.

7. Conclusion. We discussed the problem of finding dominant structures of ergodic Markov processes. Correct identification of dominant structures is very important for accurate description and complete understanding of observed processes. However, most existing methods focus mainly on reversible processes and identification of their metastable sets, as these parts of the state space often capture most interesting dynamics. However, in the case of nonreversible processes the analysis of dominant structures has to regard metastable sets as well as dominant cycles.

In this paper, we presented a comprehensive approach for finding dominant structures of Markov models for both reversible and nonreversible processes. This theory can be seen as an extension of spectral methods for identification of metastable sets of reversible processes, to the more general cases. It uses the stochastic cycle decomposition for distinguishing between different types of transition patterns and allows uncovering structures additional to metastable sets in more detail than existing approaches. We show that the additional information about cycles is essential for understanding nonreversible nature of the process, which we can then systematically describe by Schur decompositions. To demonstrate the effectiveness of our new method, we applied it to analyzing Langevin dynamics, where we successfully identified both metastable sets and cycles as dominant structures of this process, i.e. we identified metastable sets representing high and low-energy clusters as well as dominant cycles corresponding to the periodic behavior around the basins of attractions.

While our theory gives a rather complete picture of different dominant structures in Markov models, the dependence of cyclic behavior of the process on the choice of the lag time τ should not be neglected. In order to capture cyclic behavior for all important lag times, one is interested in analyzing the infinitesimal generator L of the process. Although our theory can be directly applied to processes given by the infinitesimal generator L , for many realistic models an infinitesimal generator does not exist, e.g. molecular dynamics [42, 41]. This problem needs further investigation and will be the topic for future research.

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Principle” for fruitful discussions about the need of methods for nonreversible processes.

8. Appendix. In the following it is shown that a perturbation of a 1- or 2-cycle of a reversible transition matrix with a double eigenvalue can never drive this eigenvalue into the complex plane.

Perturbation of a single entry of P . If only the entry (q, r) , $q \neq r$, of P is perturbed we have

$$L_{kj} = \sigma \delta_{kq} (\delta_{jr} - \delta_{jq}),$$

where σ is the strength of the perturbation. Let the q -th entry of u_j be a , b the r -th entry of u_j , and α the r -th entry of u_{j+1} , and β the r -th entry of u_{j+1} . Then

$$\hat{L} = \sigma \mu_q \begin{pmatrix} (b-a)a & (\beta-\alpha)a \\ (b-a)\alpha & (\beta-\alpha)\beta \end{pmatrix}.$$

Then condition (4.4) reads

$$\left((b-a)a + (\beta-\alpha)\alpha \right)^2 < 0,$$

which can never be satisfied. Thus, the perturbation of a single entry of P can never move a double eigenvalue into the complex plane.

Perturbation of a 2-cycle. When the perturbation is just affecting the 2-cycle $\gamma = (k, l)$ we have

$$L_{ij} = \sigma \begin{cases} 1 & (i, j) = (k, l) \text{ or } (i, j) = (l, k) \\ -1 & (i, j) = (k, k) \text{ or } (i, j) = (l, l) \\ 0 & \text{otherwise} \end{cases}.$$

Then

$$\langle u, Lv \rangle = u^T D L v = \sigma \left(\mu_k u_k (v_l - v_k) + \mu_l u_l (v_k - v_l) \right) = \sigma (\mu_k u_k - \mu_l u_l) (v_l - v_k),$$

Denoting $u_j = u$ and $u_{j+1} = v$ we thus get

$$\hat{L}_{12} \hat{L}_{21} = \sigma^2 (\mu_k u_k - \mu_l u_l) (v_l - v_k) (\mu_k v_k - \mu_l v_l) (u_l - u_k)$$

and

$$(\hat{L}_{11} - \hat{L}_{22}) = \sigma \left((\mu_k u_k - \mu_l u_l) (u_l - u_k) - (\mu_k v_k - \mu_l v_l) (v_l - v_k) \right)$$

so that

$$\frac{1}{4} (\hat{L}_{11} - \hat{L}_{22})^2 + \hat{L}_{12} \hat{L}_{21} = \frac{1}{4} (\hat{L}_{11} + \hat{L}_{22})^2 > 0$$

and thus condition (4.4) can never be satisfied. Thus, the perturbation of a 2-cycle can never move a double eigenvalue into the complex plane.

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