CURRICULUM VITAE

Jason de Joannis

1515 Pierce Drive, Emory University, Atlanta GA 30322
(404) 712-2983 (O), (404) 402-1332 (H)
jdejoan@emory.edu, http://reef.chem.emory.edu/~dejo

Education

Ph.D. Chemical Engineering, University of Florida, 2000
Polymer simulation, adsorption and confinement. Advisor: Ioannis Bitsanis

B.S. Chemical Engineering, Worcester Polytechnic Insitute, 1996
Thesis on bioreaction engineering

Employment

2002-present: Postdoctoral fellow
Department of Chemistry and Emerson Center for Scientific Computation, Emory University
Lipid bilayer mechanical properties; Hybrid MC/MD simulation of multi-component lipid bilayers
Advisor: James Kindt

2000-2002: Postdoctoral fellow
Max Planck Institute for Polymer Research, Germany
Ewald summation in two dimensions; Structural oscillations in aqueous polyelectrolyte layers
Advisors: Christian Holm & Kurt Kremer

1996: Computer assisted chemical engineering education
Worcester Polytechnic Institute
Fluid mechanics web course, Thermodynamics educational softare

1994: Research intern
Monsanto Company, Plastics R&D, Springfield, MA
High-grade ABS plastic bench-scale formulation and testing

Teaching Experience

- Instructor: Thermodynamics (U. Florida, 1999)
- Course lectures: Statistical Thermodynamics (Emory, 2003), Physical Chemistry (Emory, 2004), Phase & Reaction Thermodynamics (U. Florida, 1999)
- Teaching assistant: Kinetics & Reaction Engineering and Thermodynamics (U. Florida, 1998)

Publications

1. "Configurational Bias Monte Carlo Applied to Lipid Membranes in the Semi-grand Ensemble," de Joannis and Kindt, *Computer simulation studies in condensed matter physics XVI*, Eds. D.P. Landau and H. B. Schuttler (Springer Verlag, 2004).

2. "Electrostatics in Periodic Slab Geometries II," de Joannis, Arnold and Holm, *J Chem Phys*, 117, 2503 (2002).

3. "Electrostatics in Periodic Slab Geometries I," Arnold, de Joannis and Holm, *J Chem Phys*, 117, 2496 (2002).

4. "Scaling of Homopolymers Next to Adsorbing Surfaces," de Joannis, Ballamudi, Thomatos and Bitsanis, *Europhys Lett*, 56, 200 (2001).

5. "Compression of an Adsorbed Polymer Layer of Fixed Mass," de Joannis, Jimenez, Rajagopalan and Bitsanis, *Macromolecules*, 34, 4597 (2001).

6. "Homopolymer Physisorption," de Joannis, Park, Thomatos and Bitsanis, *Langmuir*, 17, 69 (2001).

7. "Interaction Between Undersaturated Polymer Layers," Jimenez, de Joannis, Bitsanis and Rajagopalan, *Macromolecules*, 33, 8512 (2000).

8. "Bridging of an Isolated Polymer Chain," Jimenez, de Joannis, Bitsanis and Rajagopalan, *Macromolecules*, 33, 7157 (2000).

9. "A Polymer Chain Trapped Between Athermal Walls," de Joannis, Jimenez, Rajagopalan and Bitsanis, *Europhys Lett*, 51, 41 (2000).

– To appear in November or December in *Macromolecular Theory and Simulation*: "Random-end-switching configurational bias Monte Carlo for long chain molecules," Karaiskos, de Joannis, Anastasiadis and Bitsanis

– Sent to *Langmuir*: "Simulations of Mixed Lipid Bicelle Systems: Composition and Line Tension of a Stabilized Bilayer Edge," de Joannis, Jiang and Kindt.

Activities

– Four invited seminars: Strasbourg, Heraklion, E. Lansing & Rolla

– Eleven conference presentations; chaired one conference session

– Attended 4 short courses and workshops (Particle Sci, Molec Sim, Poly Phys, Comp Biophys)

– Member of AIChE (American Inst. Chem. Eng.) since 1998; Society for Biological Engineering; past member of APS and ACS

References

Anthony Ladd
Professor, Department of Chemical Engineering, University of Florida, Gainesville, FL 32611, ladd@che.ufl.edu, (352) 392-6509

James Kindt
Assistant Professor, Department of Chemistry, Emory University, 1515 Pierce Dr, Atlanta, GA 30322, jkindt@emory.edu, (404) 712-1817

Christian Holm
Project Leader, Max Planck Institute for Polymer Research, P.O. Box 3148, 55021 Mainz, Germany, holm@mpip-mainz.mpg.de, +49-6131-379141

Ioannis Bitsanis
Research Professor, Foundation for Research and Technology - Hellas, Institute for Electronic Structure & Lasers, P.O. Box 1527, Heraklion 71110, Crete, Greece, bitsanis@iesl.forth.gr, +30-2810-391383

RESEARCH PLAN

JASON DE JOANNIS

The following outlines my proposals for the development of a nationally funded research program in the areas of biopolymers and molecular simulation. Specific plans are developed on the topics of ring polymers, molecular elasticity and simulation methodology.

These topics will require input from multiple disciplines which I have acquired from my education in chemical engineering and postdoctoral research in physics and chemistry departments. I will draw on experience in advanced simulation techniques such as hybrid Monte Carlo/molecular dynamics and Ewald summation in various geometries.

The topic of ring polymers is particularly intriguing. Understanding the properties of natural and synthetic variants will pave the way for this unique material in biomedical and industrial engineering applications. In Section 1 three projects are defined on charged rings, linked-ring dynamics and cellular transcription.

Section 2 is devoted to the topic of microscopic elasticity. A wealth of interesting experiments have probed the mechanics of single DNA molecules and may help create DNA motors. Two problems are defined here on DNA stretching and wormlike micelles. The last section describes my general approach to the development of a research laboratory.

1. Ring polymers

Under the topic of ring polymers I will pursue three lines of research. The first problem involves conformation and writhe of charged ring polymers. The effect of electrostatics in rings in general has not been studied and is important. The second problem will deal with the dynamics of catenanes (linked rings), particularly in networks such as those used for electrophoretic separation of DNA. The final problem is connected to one of the proposed mechanisms of prokaryotic transcription and concerns disentanglement of nascent RNA from its circular DNA parent.

Why are ring polymers rare? They are less common than linear polymers because during synthesis (or at any time) the probability for the ends of a long chain to meet is statistically negligible. In spite of this obstacle, natural ring polymers abound in the DNA of bacterial organisms (prokaryotes). Recent advances [1, 2] in ring synthesis promise an opportunity to test long-standing theoretical ideas developed before the existence of rings was known.

There are several good reasons to study ring molecules. Not least of these is their central role in the cellular metabolism of prokaryotes. Many questions arise ranging from entanglement alluded to above, to cellular evolution, to the effects of supercoiling on genetic packaging and access. Ring molecules also offer a unique engineering material for nanotechnology and advanced materials. A pair of interlocked rings has been used [3] as an electrically operated nano-switch. Another application is the creation of "Olympic" gels or rubbers without chemical cross-links. Finally rings

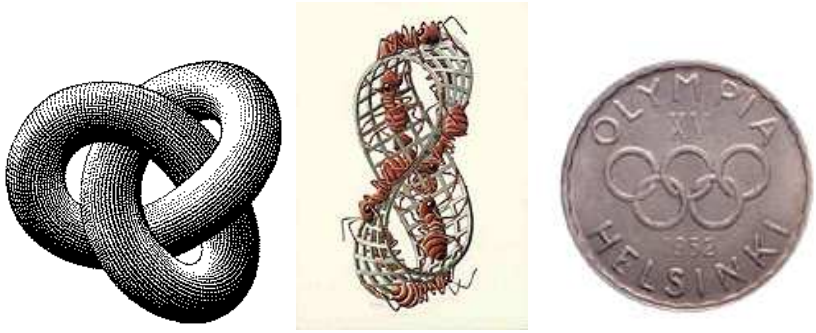

FIGURE 1. Artistic renderings that illustrate the three topological classes that arise in rings: knots, topoisomers and catenanes (left to right).

offer an ideal platform from which to isolate the effect of topology on polymers and random walks in general, which impacts many other areas such as Brownian motion, magnets and electronic structure.

In spite of the simplicity of the idea of closing a linear polymer into a ring the consequences can be dramatic. The lack of chain ends may serve to protect bacterial DNA from degradation [4], collapses rings in the melt phase [5] and changes the fundamental mechanism of diffusion [6]. We are only beginning to explore the effect of topological constraints on the properties of ring polymers. Figure 1 shows the three kinds of topology that can arise in rings. The relative frequency of entanglement varieties depends on the conditions of synthesis [7]. We can expect concentrated conditions to favor catenation (interlocking rings) for example. The coin in Figure 1 shows a linear pentameric catenane.

Knots such as the trefoil shown in Fig 1 at the left, can induce collapse or suppress solvent effects [8] (not unlike a bimetallic strip) depending on their kind and density. Knot theorists took an early interest in the dependence of the knotting probability on ring size if all conformations are weighted equally. This problem remained largely intractable until the advent of computer simulation, when the probability was found to approach unity for large chain lengths [9]. Therefore knots should be expected in DNA unless synthesis is guided.

In double stranded ring polymers torsional topology is introduced. The molecule can be visualized effectively as a ribbon as in Fig 1, which can have any number of twists before closure. These are called topoisomers distinguishable by their linking number (number of twists). A large linking number produces a buckling effect or supercoiling as can easily be demonstrated with a rubber tube. Considerable progress has been made in electrophoretic characterization of DNA supercoils [4].

Nature engineered some impressive features into the cell. One of these is the feedback regulation of supercoiling. The enzyme gyrase transiently nicks one strand and increases the linking number. This in turn inhibits the recombinatary production of gyrase, acting essentially as a chemostat. The abundance of supercoiling implies it has important biological functions. It may help to produce chromatin, an accessible, organized and compact package for the genetic code. It may also promote protein and sequence recognition as well as enhance mobility.

Models for bacterial metabolism must specifically account for the possibility of entanglement during the synthesis of new DNA, RNA or protein molecules. These cooperative processes are known as replication, transcription and recombination respectively. One imagines an enzyme (or an ant) that walks along the helical groove of the template-DNA, un-zipping each locality and trailing an entangled nascent molecule. One possible outcome is a pair of multiply catenated rings that then require an enzyme mediated de-catenation. There is much to be learned about these processes.

Charged Rings. The effect of charges on ring polymers has not been studied beyond the Debye-Hückel level. This is especially relevant to DNA which is a highly charged molecule. Electrostatic interactions, because of their long range, are known to have unusual consequences in complex liquids; examples include Debye-Hückel theory, attraction between like-charged colloids [10], viral-DNA condensation [11] and polyelectrolyte swelling in super-absorbents.

The general effect of charges is to swell the conformations of a polyelectrolyte since its backbone contains only like-charges. When more concentrated solutions are considered, multiple length scales and a complex phase diagram appear [12]. Open polyelectrolytes scale linearly in size with number of monomers and assume extended cigar-like conformations. It is unknown whether ring polyelectrolytes have the same scaling and they may have a more sphero-symmetric or circular shape with larger fluctuations since all momomers in a ring share the same distribution function. If knots are present, they can be distributed throughout the chain or localized by external tension or internal long-range forces [13].

I plan to use Monte Carlo methods to explore the configuration space of a charged ring. The use of a lattice and the bond-fluctuation model will allow tabulation of distances for efficient pair interactions. We will test several promising moves for sampling efficiency such as vector shuffling [14], pivot-coupled GCMC [15], internal reptation, directed configurational bias [16] and dimerization [17]. These moves will freely allow the crossing of the backbone in principle sampling all types of knots. Most sampling will occur in the simple knot types, which will be distinguished computationally using the appropriate Alexander polynomial [9].

In supercoiled DNA the effects of the buckling stress on structure are quantitatively characterized by "writhe" (i.e. tortuosity of path) as well as distortions in the helical geometry itself, known as "twist". Knowledge of the writhe distribution can be used to infer the torsional modulus of DNA from electron micrographs [18]. Our Monte Carlo simulations will provide the first information on writhe in charged rings with a linking number of zero.

It is useful to compare conclusions drawn from independent methods and/or models to provide a broader perspective. An alternative approach is Poisson-Boltzmann theory which yields useful analytical asymptotes. Several simplifications are usually required including linearization of the distribution function, neglecting excluded volume or solvent effects and ignoring counter-ions (i.e. infinitely dilute system). I will use the cell model [19] to investigate the counter-ion distribution in two geometries. The first is a disc-shaped cell encompassing an entire ring. The second is related to a common supercoiled structure that occurs in buckled topisomers, the plectoneme (pictured at right). We will model the middle of a very long plectoneme with a cylindrical cell that periodically repeats in the axial direction. This will be compared to one-dimensional Ewald

simulations. These comparisons will provide quantitative bounds for the linearization, infinite-dilution and solvent approximations as well an analysis of rings at a different scale.

This project will also provide valuable experience in dealing with the technical issues of equilibrating ring molecules effectively. Some effort will be dedicated to the issue of constant-topology simulation. How can we efficiently sample the configurations of, for instance, an un-knotted ring? The obvious answer is to use a method that mimics real dynamics such as stochastic dynamics or bond-fluctuation Monte Carlo, but there may be better alternatives. Constant topology simulations will become particularly important as many-chain systems are targeted, which is a long-term goal.

Catenane Diffusion. The dynamics of rings has received considerable experimental and theoretical attention. Ring diffusion was expected to be exponentially slow as in branched polymers. Instead observations show a power-law scaling of the diffusion constant that is similar to linear polymers. Thus far no equivalent entanglement length has been found for ring polymers [20], which for linear polymers dictates the crossover to the reptation regime. The most prominent model for diffusion makes an analogy to a transiently branched chain [6]. The slow diffusion observed in mixtures of open and closed chains [21] has not been explained. This is a property that could be exploited with telechelics for applications requiring reversible changes of viscosity.

I will investigate the dynamics of catenanes. A comparison will be made between the diffusion coefficients of a single ring, a pair of linked rings, and higher order catenanes. This will be conducted using stochastic molecular dynamics. To ensure that the rings do not become unlinked, finitely-extensible-elastic-springs will be used to prevent bond crossing. I will be interested in the self-diffusion of catenanes as well their driven diffusion in an electric field of fixed strength.

Of considerable interest will be the development of a fixed network for the chains to diffuse through as in experimental gel electrophoresis. The chief question is the network diffusion dependence on the catenation number. These results will hopefully prompt experimentalists to refine techniques of catenane production in order to conduct similar experiments and develop catenane nanotechnologies. Moreover, this information will help distinguish catenated bacterial DNA from its counterparts.

RNA-polymerase Transcription. RNA-polymerase is the enzyme that carries out transcription - the creation of an RNA molecule from a DNA template. The enzyme must sequentially access the information contained in the base pair sequences that straddle the double helix. The question remains, how is permanent topological entanglement between the two molecules avoided? Several proposals exist to explain this process. In one model the enzyme travels along the spiral groove of DNA, un-zipping, reading and re-zipping each part as it advances. On the other hand the model which I intend to examine, proposes that the enzyme is stationary and causes a torque on the local section of DNA to which it is attached. This torque twists the DNA like a motor and causes it to circumnavigate.

To account for the effect of twist in a simple bead-spring chain, we will introduce an anisotropic torsional potential for the dihedral angle. The external twisting effect will be represented by a small cylinder fixed in space and having the same length

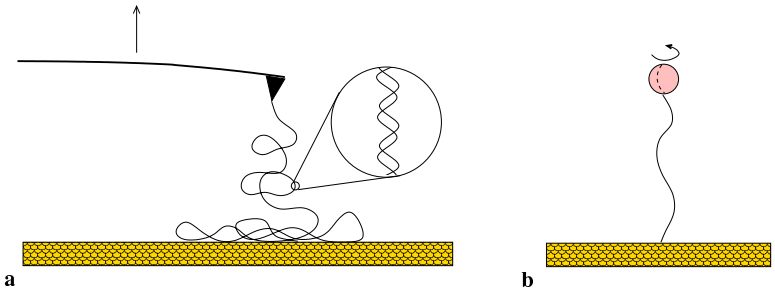

FIGURE 2. Elasticity experiments. a) AFM stretching of DNA, b) Torsional elasticity - magnetic bead.

as the distance between bonds. Any bead within the tube can only exit through the ends and experiences a constant torque or twist energy.

This will yield information about the range of torques that can produce circumlocution and the corresponding rates. Moreover I will study the effect of linking number as an inhibitor or facilitator of this process. Supercoiling may create resistance to twist-induced motion. On the other hand, it often produces ordered structures whose qualitative effect is more difficult to predict.

2. Elasticity

This section defines two projects related to microscopic, or molecular elasticity. Both resemble the classical worm-like chain proposed many years ago by Kratky & Porod. The model was meant to correct for the stiffness of DNA, which has the longest persistence length known. At the level of a random walk, all polymers have in common an "entropic elasticity" understood as a consequence of the strain-induced shrinkage of configuration space. This contributes a linear, Hookean resistance to all forms of strain whether it is confinement or stretching.

Biopolymers. The double helix also has an inherently anisotropic torsional elasticity. The local bending is influenced by composition since some base pair sequences have an intrinsic curvature along their helical axis. This may help sequence recognition by localizing them to strongly bent apical loops in supercoiled structures.

Experimental manipulation of single biomolecules such as titin and DNA has been performed with the atomic force microscope (AFM) as well as magnetic and optical tweezers. These procedures are illustrated in Fig 2. Since biopolymers are much longer than synthetic ones – the length of a DNA molecule containing the human genome would measure two meters if stretched out – single-molecule experiments with biopolymers can realize the infinite chain-length assumption so ubiquitous in polymer physics.

A recent experiment [22] provides an example of the increasing sophistication of single-molecule mechanics. A magnetic particle is manipulated to build up torsional forces in the DNA molecule which are then released through a coupling to a colloidal particle. The motion of the colloid is captured with video microscopy and provides information on the molecule's torsional modulus.

DNA stretching experiments have identified four major regimes spanning a range of forces from 0.01 to 10,000 pN [23]. The two regimes of weakest response, known

as the entropic and intrinsic elasticity regimes, are dominated by the universal features of random walks. The final two regimes under higher loads are marked by helical distortion and bond rupture.

My goal will be the development of a DNA model that incorporates enough of the helical structure to describe the intrinsic elasticity regime. Current models include a double stranded worm-like chain, a triangulated ribbon, a string of rods, a string of ellipsoidal beads and deformable tubes [24]. These hope to capture some of the helical or ribbonlike nature of DNA and attempt to navigate the tradeoff between computational efficiency and geometrical specificity. I will account for long-range electrostatic forces more accurately than in worm-like chain field theory. The system will propagate by Langevin dynamics using the Technetium package. This was developed in our laboratory by Y. Bouret (now at ENS-Paris) and it uses a library of coarse-grained building blocks: various kinds of beads, cylinders and off-center bonds. Locally, a well-formed helix will result from use of a specialized torsional potential through the dihedral angles of a bead-spring chain [25].

The confinement of biopolymers is also marked by an elastic response of the same kind. For instance three-dimensional confinement occurs in viruses where a DNA molecule is tightly packed inside a hard protein capsule. Two-dimensional confinement can occur at the surface or within cell membranes due to adsorption or encapsulation inside rafts. I plan to study single and multiple polyelectrolyte confinement problems in two and three-dimensional geometries. This will be accomplished using the parallel molecular dynamics package ESPRESSO developed at my former laboratory in Mainz, which focuses on an efficient parallel implementation of Fast Fourier methods for electrostatics.

This work on stretching and confinement will be complemented by a phenomenological scaling analysis along the lines of polymer-blobs. This type of analysis captures the more general features of conformational distortion and led to integration of the processes of polymer adsorption, stretching and confinement under a single concept [26].

Worm-like micelles. Lipid assemblies are an essential part of cellular systems and commonly form bilayers, vesicles, worm-like micelles, spherical micelles and bilayer-micelles. Bilayers produce a membrane of high mechanical strength and flexibility useful for encapsulating systems in an aqueous environment. The mesoscopic Helfrich model provides the framework for description of elastic membranes and has been used for instance, to study wrapping of a membrane around a colloidal particle. Experimental and simulation techniques have been refined to provide estimates of the material parameters in the Helfrich model, including the bending modulus and intrinsic curvature of the bilayer surface, the surface tension and the line tension and modulus of a bilayer edge.

In this project an atomistic simulation of a worm-like micelle will be used to study its effective mechanical properties. The GROMACS molecular dynamics package will be employed using the phospholipid/solvent force-field which has reproduced many bilayer properties. This package is easily extended and will be modified to include a one-dimensional Ewald summation. The spectrum of fluctuations of the worm-like micelle axis will provide for a direct calculation of its bending constant under zero line tension. This will be corroborated with other mesoscopic methods of analysis. The pressure coupling of the system can be adjusted to induce a positive or negative line tension producing buckled and stretched conformations, producing

a broader range of conditions for mechanical modeling. Worm-like micelles have been implicated in a proposed lamellar phase punctuated by bridging stalks and a branched worm-like micelle phase [27]. Preliminary progress has been made on the construction of stable starting samples for these systems as a biproduct of my work on bilayer micelles.

3. Roadmap

Accomplishment of research goals outlined above requires a well-defined plan for the development of laboratory and computational resources and the cultivation of a knowledgeable and productive group. The establishment of a mature research laboratory including funding, equipment and students will take several years. I plan to develop a computational cluster, obtain funding from NSF and NIH and develop supportive and productive relationships within the department and within my group.

Early on it is crucial to focus on the establishment of a funded research program. I have had some exposure by attending an AIChE workshop given by G. Prentice - director of the CAREER award - and by applying for funding from the NIH Institute of General Medical Sciences. To improve my awareness of good proposal writing I will to seek advice from colleagues in the department, build perspective on the key questions in my field through contacts from conferences, seminars and correspondence. As a time management strategy I will schedule *regular* time for uninterrupted scholarly reading and proposal writing and make sure not to exceed budgeted time for course lecture preparation.

The laboratory requirements consists principally of a 16-20 node, parallel computing cluster. The cluster will run on open source software with off-the-shelf PC components that can be redeployed as workstations and easily extended or upgraded. I have had experience in cluster building and management as administrator in my current lab and through the computational biophysics summer school at UIUC. The target is to install and benchmark MPI, FFT and GROMACS software within a couple of weeks. Quotes from Microway, Atipa and Penguin including a Gigabit communication switch are around $50K. I will require well air-conditioned laboratory space to maintain this system.

Students will initially receive well-defined problems than can be tackled quickly and learned from. As they mature, some flexibility will be offered in the direction they wish to pursue within the above research areas. I will encourage a team-oriented culture within the group through meetings and traditions. I am interested in getting my students involved with a shared-facility AFM and/or collaborating with experimentalists who are using optical tweezers or AFM.

At the entry of professor's career I recognize that it is important to make a determined and rapid start. I intend to set goals and priorities and periodically review and share these with other faculty members and the department chair.

References

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