

3.942 (Polymer Physics) Notes

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Topic 0

Introduction

0.1 Introduction

9/4:

- Starts at MIT time.
- Alexander-Katz begins.
- Extra notes on the syllabus.
 - All of the numbered courses were combined recently because offerings were almost identical.
 - Alexander-Katz is a DMSE prof; Doyle is a ChemE prof.
 - On textbooks.
 - He used to send us to the library, and we'd sort out which text was easiest for us to understand.
 - He recommends we buy Lodge and Hiemenz (2020), but the chapters will also be posted.
 - The first chapters in the book are chemistry, and the later ones are physics.
 - Strobl (1997) will give us more information about some areas. Good if you're interested in mechanics, solid states, and plastics.
 - Young and Lovell (1991) is similar to Lodge and Hiemenz (2020).
 - Rubinstein and Colby (2003) is advanced undergrad to grad, and has lots of examples. Good for understanding scaling concepts. Doesn't have any hard phases. Has a bit on characterization methods.
 - de Gennes (1979) is widely hated because it was written very colloquially, but the ideas are really interesting. The man himself had a very straight mind, and thought about problems in a very interesting manner.
 - There will be about 6 homeworks, 1-2 per quiz.
 - They will give us a key, and we will have to grade our own homework.
 - We will then have to turn in a second homework, which is revised and has a grade on it.
 - In Alexander-Katz's experience, this method gives a 10% increase in quiz scores on average. This is because going through your homework in depth is very beneficial!
 - Also, the last few times they had a grader, (s)he usually knew nothing about polymer physics.
 - There will be 3 quizzes.
 - Each counts for 20%; they're not cumulative.
 - Alexander-Katz and Doyle will be grading our quizzes themselves.
 - Make sure to participate! Don't stay quiet; ask questions, etc.
 - You may work together; just note your collaborators' names on your assignments.
 - Don't trust ChatGPT and generative AI 100%. You won't be able to use them for the quizzes, but they may be useful for searching.

- Lecture now begins.
- **Hard matter:** Metals, ceramics, and semiconductors, which are typically highly crystalline.
 - For hard matter, the binding energy ε between atoms is much greater than $10^{-21}k_{\text{B}}T$.
- In this course, we're going to be working with materials of $\varepsilon \approx 1 \text{ eV} \approx 10^{-19} \text{ J}$.
 - So to break these bonds, it takes about $100k_{\text{B}}T$.
- **Soft matter:** Polymers, organics, liquid crystals, gels, foods, life!
 - $\varepsilon \approx k_{\text{B}}T$.
- A good mental picture for the barrier dividing small molecules and soft matter is the following.
 - Ethane is a gas. Very little forces between molecules.
 - When we make polyethylene, it can become extremely strong. This is because of the **multivalent effect**. When the chains are long enough, we get a solid.
- **Multivalent effect:** The observation that multiple copies of a weakly binding molecule — when arranged on a common scaffold — exhibit a significantly stronger and more specific binding response than a single, isolated molecule.
- A fundamental idea: As the **degree of polymerization $n = N$** grows, a lot of interesting behavior appears.
 - In this course, most of the behavior we're concerned with will be *qualitative*.
 - For example, we don't care about *exact* numerical properties if $n = 64$, but we do want to answer questions like, “if n doubles, what will happen?”
- Topics to understand in this class (the key origins of soft matter behavior).
 - Relatively weak forces between molecules.
 - We'll discuss this today.
 - Various types of bonding.
 - Molecular shapes and sizes.
 - What chemistries are relevant.
 - Fluctuating molecular conformations/positions.
 - Influence of solvent, diluent, particles, surfaces.
 - Entanglement.
 - Alexander-Katz gives the pasta analogy.
 - Entanglement gives interesting rheological properties, dramatic increases in viscosity (much higher than you'd expect from simple arguments), etc.
 - Many types of entropy.
 - What architectures and structural hierarchies exist over several length scales (nano, micro, etc.).
- We'll also discuss some characterization methods for harder and softer materials.
 - We'll do the theory first, and then figure out what to measure to figure out what's in the materials.
- Lastly, we'll talk about some opportunities to exploit the properties of soft matter in diverse applications.
- Aside: They used to have a final project on how to make contact lenses comfortable.
 - They are semisoft and silicone-based nowadays!

- Two types of interactions in soft matter: **Intramolecular** and **intermolecular** interactions.
- **Intramolecular** (interactions): (Macro)molecules are predominantly held together by strong covalent bonds with many conformations accessible via rotational isomeric states.
 - Think Newman projections. These reduce the possible conformational states substantially.
 - The allowed conformations are **gauche plus**, **gauche minus**, and **trans**. This reduces the polymer entropy a lot.
 - We will talk about how this tacticity affects polymer size and other quantities.
- **Intermolecular** (interactions): Hard sphere, Coulombic (including stacking; usually just a $1/r$), Lennard-Jones (induced dipoles, such as van der Waals forces), or hydrogen bonding (net dipoles; polar interactions will definitely be important).
 - We think of polyethylene as not that polar, but it is kind of polar (especially if you put a methyl group at every other position, as in polypropylene).
 - The PE vs. PP T_g 's vary *dramatically*.
- **Short range order**: When you turn molecules into polymers, other length scales start to matter. *Also known as SRO*.

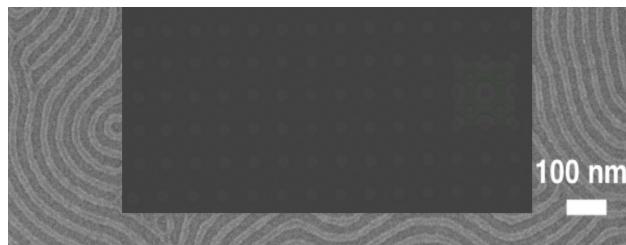


Figure 0.1: Short range order in PS-*b*-PDMS.

- Consider a block copolymer of polystyrene and PDMS. When you put it all together, it forms a fingerprint-like pattern (PDMS light, PS dark).
- The chains look random at the nanoscale, but they phase-separate when you zoom out a bit.
- Think of this as spaghetti on a surface.
- SRO is always present in condensed phases.
- **Long range order**: Visible in perforation lamellae, for example. *Also known as LRO*.
 - LRO is sometimes present in condensed phases.
- Example: If a material is ordered by *layer* from bottom to top, defects can assemble and give you color control. This happens in some soft matter films.
 - In the specific example in the slides, P2VP keeps a charge always, and polystyrene does not.
 - Defects allow swelling to happen much faster, and swelling changes length scales which changes color (almost like in quantum dots, where size affects color).
- Overall, the goal of soft matter studies is the manipulation of structures, orientation, and defects. We seek to develop methods to process polymers and soft solids, so as to create controlled structures and hierarchies.
- Mantra: Whatever we want to do with polymers, nature has done it first.
 - Indeed, the color changing thing above is the same principle as color-changing octopi!

- History of polymers.
 - First users were in Mesoamerica (around 1600 BC). They harvested natural latex (polyisoprene) from trees. As soon as it comes out, it hardens. They used this polymer to make a game ([prehispanic basquethol](#)) and shoe soles.
 - If you don't crosslink latex, it will be like silly putty, going everywhere.
 - They crosslinked it with the juice of a cactus that they had chewed.
 - 300 years after the Spanish invaded (around 1850 AD) and crosslinking was forgotten, Goodyear figured out once again how to crosslink natural rubber (he did so with very strong acids, sulfuric acid and such).
 - Aside: Humans tend to find uses for materials far before they understand what it's made of, how to process it, etc.
 - Pretty much all of biology is made out of polymers: DNA, RNA, proteins, cells, etc.
 - People only discovered polymers (in the modern sense) in the 1920s, when H. Staudinger put forth the **macromolecular hypothesis**.
 - After Staudinger, people started thinking about what they would *like* to make.
 - 1930s: Silk was the highly desired textile of the time.

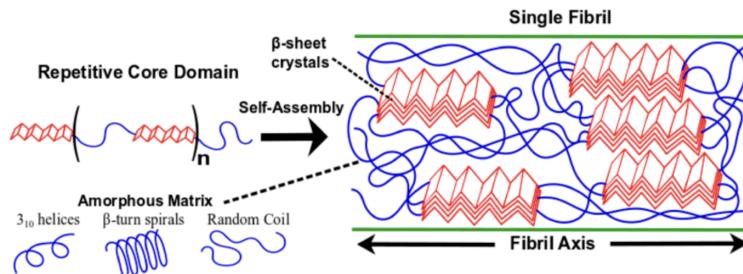


Figure 0.2: Silk.

- Made out of proteins with amorphous and crystalline (β -pleated sheets) regions (Sarkar et al., 2019).
 - So they thought about repeating a peptide (polyglycine). Even more similarly, though, Carothers (really talented chemist who went to Dupont) made nylon out of alternating aliphatic segments and hydrogen-bonded amides!
 - Nylon is made via **step-growth polymerization**; Alexander-Katz diagrams this out on the blackboard.
 - Nylon has very nice properties and is even stronger than silk. Nicer properties than silk.
 - Changing the number of carbons in the chain varies the melting temperature!
 - Note that members of the nylon family are named by counting the number of carbon atoms in the backbone between nitrogen atoms.
 - 1965: Stephanie Kwolek (also Dupont) develops Kevlar.
 - Better hydrogen bonding, much stiffer.
 - Same number of carbons as nylon 6! It really does matter how you arrange the carbons.
- Common polymers.
- **Polyethylene:** *Also known as PE.*
 - Invented 1933.
 - One of the biggest polymers on the market today.

- Polyester.

- Invented 1941.
 - You can make many different kinds of these; defined by the ester linkage.
 - Nature is full of polyesters, all very different.
 - If you change one carbon on an oligoester, it can go from tangarine-sented to very bad.
 - Used for lubrication, since the polar carbonyl moiety gives interesting properties.
 - These don't have the N–H moiety of polyamides, but can still be assembled by condensation.
 - These melt very easily.
 - Example: Polyethyleneterephthalate, PET.

- Common classes of polyolefins.

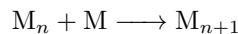
- Vinyl polymers, diene class, vinylidenes.
 - Teflon (polyvinylidene fluoride).
 - Plexiglass: Methacrylics.
 - Acrylics are more liquid; methacrylics are harder and more solid.

- Common classes of step-growth polymers.

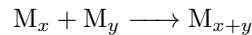
- Silicone.

- Polymer nomenclature.

- Addition reactions.



- Condensation reactions.



- **Monomer:** Small molecule.
 - **Oligomer:** $n < 100$.
 - **Polymer:** $n \in [100, 10000]$.

- Characteristic properties of polymers (depend on the chemistry, *and* how everything is arranged).

- Insulating or conducting.
 - Light-emitting.
 - Photovoltaic, piezoelectric.
 - Soft elastic (really large reversible deformations, often temperature dependent like the PGS rubber band example) or very stiff.
 - Zero/few crystals or highly crystalline.

- Polymers are in everything now.

- About 50% of a plane is made out of composites (like carbon fiber; very strong and very light).
 - It's really expensive to cure the polymers that hold all the carbon fibers, so they're looking for new ways to do that (this is like REMAT stuff!).

- Biggest shares of the polymer market.

- PE (plastic bags), PP, PET (clothes and garments), PVC (pipes), and polyamides.
 - They are trying to make the polymer market “look” better now by calling it the “*circular* polymers market.” Alexander-Katz doesn't believe properties are the same in recycled polymers.

- Thermodynamics refresher.
 - Given your state variables (T , P , E , s , etc.), some states are allowed to a system and some are not.
 - We usually work with the **Helmholtz free energy** in this class, not the **Gibbs free energy**. Alexander-Katz doesn't care about the difference; Doyle will.
 - If you start in an out-of-equilibrium state, thermodynamic forces will push you down into an equilibrium state. We will think of forces as the derivative of the slope of the energy hypersurface.
- Statistical mechanics primer.
 - We will use Boltzmann's law, $S = k \ln(\Omega_{\text{tot}}[U])$, a lot.
 - Because suicide is so stigmatized, his tomb (in Vienna, Austria) is very hidden away.
 - In the nineteenth century, there was a big debate over whether the world was molecular or continuous. Chemists (who tend to be right) said molecular, and physicists said that we didn't need to worry about that. Boltzmann tried to reconcile these two philosophies.
 - He eventually discovered the following. Ω_{tot} is all the possible ways you can arrange the system. Technically, $\Omega_{\text{tot}}[U]$ is the total number of equivalent microstates at constant energy, U . If you take the natural log and multiply by a constant, we get the entropy.
 - Recall that Boltzmann's discovery can be related to the pressure of ideal gases.
- Next Tuesday, Prof. Doyle will begin on single chains.

Topic 1

The Macromolecule

1.1 The Macromolecule

9/9:

- Pat Doyle begins; he will teach the next three lectures.
 - They've broken the class up into modules taught alternatingly.
 - Aside: Alfredo has taught this course 10 times; Doyle never has (he's replacing Greg Rutledge this year).
- Announcements.
 - Slides and homework 1 have been posted.
 - Slides should be posted before lecture, but may not be posted much before.
- Lecture outline.
 - Connectivity: Thermodynamic limit, architecture, and molecular weight.
 - Configurations: Structural, chemical, stereo, and geometrical isomerism.
 - Conformations: Rotational isomeric states.
- **Connectivity:** The joining of small parts (monomers) into larger molecules (polymers).
- **Degree of polymerization:** The number of repeat units in a polymer. *Denoted by N .*
 - Derivable from the molecular weight(s).
- Example: Polyethylene.
 - In this course, vinyl examples will be our workhorses, but we will “riff off of them” to other polymer types.
 - The **repeat unit** here is CH_2CH_2 , consistent with the IUPAC nomenclature of polyethylene.
 - Ethylene is also the monomer.
 - The end groups do look different, but the **thermodynamic limit** addresses them.
- **Repeat unit:** A part of a polymer whose repetition would produce the complete polymer chain (except for the end groups) by linking the repeat units together successively along a chain.
- **Thermodynamic limit:** The finding that as $N \rightarrow \infty$, the end group chemistry matters less. *Also known as polymer limit.*
 - The thermodynamic limit is also sometimes discussed in the context of statistical mechanics, where collective behavior also matters more than individual or picoscale.

- **Glass transition temperature:** The temperature at which a substance will go from brittle to kind of rubbery. *Denoted by T_g .*
- **Flory-Fox correlation:** The simple model that the T_g of a polymer asymptotically approaches a limit $T_g(M \rightarrow \infty)$ for higher and higher molecular weights at some empirically derived rate A . *Given by*

$$T_g(M_n) := T_g(M \rightarrow \infty) - \frac{A}{M_n}$$

- Alkane series often obey this simple $1/x$ relation.
- Another example of thermodynamic limits: Regardless of polymer structure, a power law defines polymer viscosity.

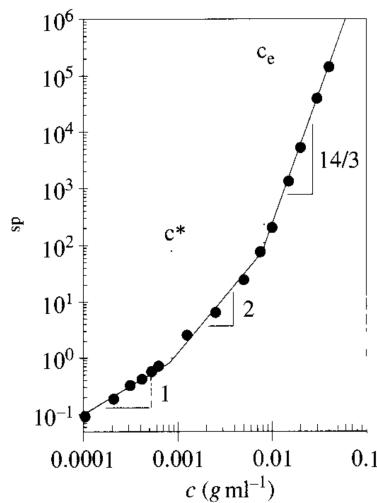


Figure 1.1: Polymer viscosity scales universally by power laws.

- Namely, polymer viscosity increases up to a critical concentration c^* at a slope of 1, then to an entanglement concentration c_e at a slope of 2, and then at a slope of $14/3$ past that.
- Thus, there are three universal scaling regimes.
 - A log-log plot is used to show power-law scalings, like in high school trigonometry!
 - Aside: Power laws are present everywhere once you get big enough, down to the volume of our lungs relative to our bodies in bigger and bigger animals.
- This class isn't super stringent on nomenclature, but it's good to know terms for when we read papers (Table 1.1).
 - Alfredo will talk about block copolymers a good bit!
- Polymer architectures.
 - Linear polymers.
 - Polyrotaxanes and other supramolecular assemblies can have interesting properties. Example: Catenated DNA!
 - Cross-linked systems (nice gelation).
 - Branched polymers can have a single monomer, or multiple as in graft (Table 1.1).
 - Dendritic polymers have different generations with regular branching for very dense structures.
 - There are a few more classes, as well.

Copolymer type	Arrangement of monomeric units	Representation	Connective	Example
unspecified	unknown or unspecified	(A- <i>co</i> -B)	- <i>co</i> -	poly[styrene- <i>co</i> -(methyl methacrylate)]
statistical	obeys known statistical laws	(A- <i>stat</i> -B- <i>stat</i> -C)	- <i>stat</i> -	poly(styrene- <i>stat</i> -acrylonitrile- <i>stat</i> -butadiene)
random	obeys Bernoullian statistics	(A- <i>ran</i> -B)	- <i>ran</i> -	poly[ethene- <i>ran</i> -(vinyl acetate)]
alternating	alternating	(AB) _n	- <i>alt</i> -	poly[(ethylene glycol) ^a - <i>alt</i> -(terephthalic acid)]
periodic	periodic with respect to at least three monomeric units	(ABC) _n (ABB) _n (AABB) _n (ABAC) _n	- <i>per</i> -	poly[formaldehyde- <i>per</i> -(ethene oxide)- <i>per</i> -(ethene oxide)]
block	linear arrangement of blocks	-AAAAA-BBBBB-	- <i>block</i> -	polystyrene- <i>block</i> -polybutadiene
graft	polymeric side chain different from main chain ^b	-AAAAAAAAAA- B B B B B 	- <i>graft</i> -	polybutadiene- <i>graft</i> -polystyrene

^a ethane-1,2-diol, ^b main chain (backbone) is specified first in the name.

Table 1.1: IUPAC nomenclature of copolymers.

- We'll now discuss some nomenclature on molecular weight.
- Aside: Other than nature, synthetic chemists cannot make dispersity 1 polymers; “polymer chemists aren’t gods, despite some thinking they are.”
- ***i*-mer:** A segment of a polymer with degree of polymeriation *i*.
- ***M_i*:** The molecular weight of the *i*-mer. *Given by*

$$M_i := i M_0$$

- ***M₀*:** The molecular weight of the repeat unit in a polymer chain.
- ***n_i*:** The number of *i*-mers.
- **Number fraction** (of an *i*-mer): The probability of picking an *i*-mer out of solution when picking a chain. *Denoted by *x_i*. Given by*

$$x_i := \frac{n_i}{\sum_i n_i}$$

- **Weight fraction** (of an *i*-mer): The probability that a repeat unit picked out of solution belongs to an *i*-mer. *Also known as weight fraction. Denoted by *w_i*. Given by*

$$w_i := \frac{n_i M_i}{\sum_i n_i M_i} = \frac{n_i (i M_0)}{\sum_i n_i (i M_0)} = \frac{i n_i}{\sum_i i n_i}$$

- Note that *i n_i* is the total number of monomers in the *i*-mer, and $\sum_i i n_i$ is the total number of monomers in solution.
- **Number-average molecular weight:** The arithmetic mean of the molecular masses of the individual macromolecules. *Denoted by *M_n*. Given by*

$$M_n := \sum_i x_i M_i = M_0 \cdot \frac{\sum_i i n_i}{\sum_i n_i}$$

- **Weight average molecular weight:** A measurement of molecular weight that gives more contribution to higher-weight molecules. *Denoted by M_w . Given by*

$$M_w := \sum_i w_i M_i = M_0 \cdot \frac{\sum_i i^2 n_i}{\sum_i i n_i}$$

- Both M_n and M_w look like moments of a distribution (i.e., first and second moment).
 - We could generalize even more, but we don't need to.
 - However, to figure out if we have a tight or wide distribution, we often look at ratios of our moments. This leads to the following definition.
- **Dispersity:** A measure of the breadth of the distribution of fragment molecular weights in a polymer sample. *Also known as polydispersity index, PDI. Denoted by D . Given by*

$$D := \frac{M_w}{M_n} = \frac{\text{second moment}}{\text{first moment}}$$

- In Lodge and Hiemenz (2020), they also derive the **variance**. You don't need to worry about the math, though.

- **Variance:** Another measure of the breadth of the distribution of fragment molecular weights. *Denoted by σ^2 . Given by*

$$\sigma^2 := M_n^2[D - 1]$$

- **Schultz-Zimm distribution:** An idealized mathematical model for polymer molecular weight distribution.

- With these definitions, we can now do homework problem number 1!
- Experimental techniques to measure molecular weight.
 - Size exclusion chromatography.
 - Osmotic pressure.
 - End group analysis.
 - Light scattering techniques.
 - More sensitive to higher moments.

- We now move onto polymer configurations.

- **Configurations:** The way things are connected or bonded together.

- Physicists and chemists have many competing definitions of “configurations,” but the one above is what we'll use in this class. Essentially, think of it as a synonym for constitutional isomerism.
- Under this definition, you have to *break bonds* to create a new configuration.
- We are *not* yet talking about rotamers (what we'll call **conformations**). As you make your polymers longer and longer, the conformational space you can explore gets bigger.
- There are 3 main types of structural isomers (i.e., configurations): **Positional**, **stereo**, and **geometric** isomers.
- **Positional** (isomers): Changing connectivity.
- **Stereoisomers:** Related to chiral centers.
- **Geometric** (isomers): Related to double bonds.

- More on positional isomers.

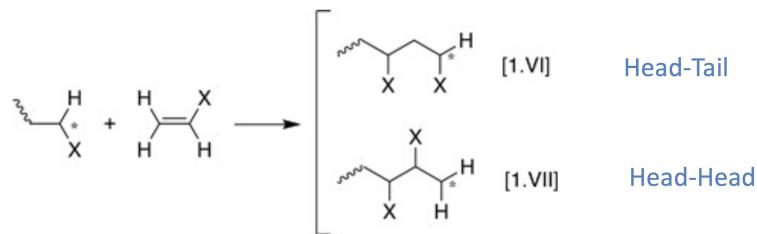


Figure 1.2: Head-head and head-tail monomer addition.

- **Head-head** vs. **head-tail** bonding in vinyl monomers.
- Head-tail is more common, and differences can change the T_g substantially.
- **Head-head** (orientation): Monomer addition wherein the substituted carbon attaches to the growing chain end. *Also known as head-to-head.*
- **Head-tail** (orientation): Monomer addition wherein the unsubstituted carbon attaches to the growing chain end. *Also known as head-to-tail.*
- More on stereoisomers.
- Doyle reviews **chirality**, **rectus** vs. **sinister**, and the **Cahn-Ingold-Prelog nomenclature**.
- **Pseudochiral** (center): A chiral center where two of the substituents are identical *except* for chirality.
 - In this case, CIP nomenclature gives priority to the branch with more R chiral centers.
- Chiral centers in polymers give rise to **tacticity**.

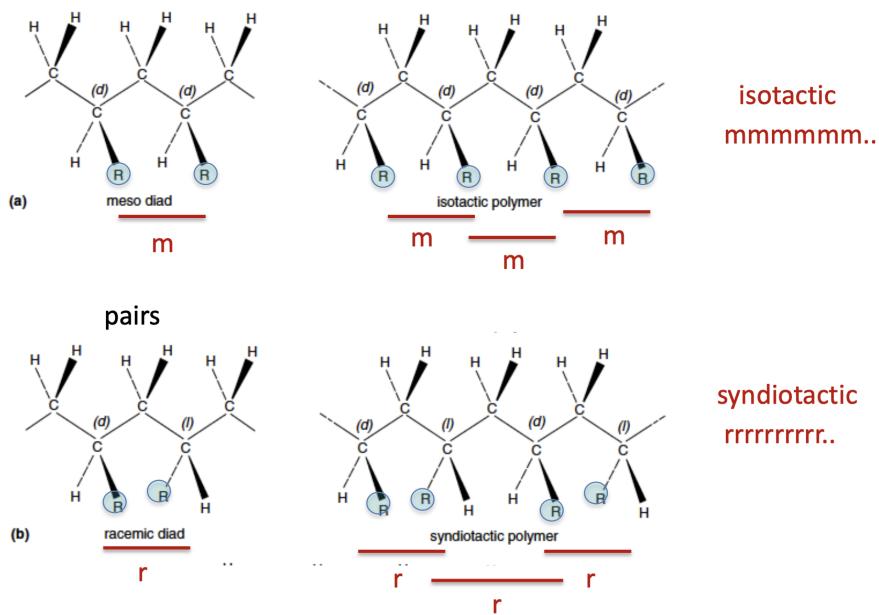


Figure 1.3: Tacticity.

- There are **meso diads** and **racemic diads**; which ones you have determine if the polymer is **isotactic**, **syndiotactic**, or **atactic**.

- **Meso** (diad): Two adjacent chiral centers with a local plane of symmetry halfway between them.
- **Racemic** (diad): Two adjacent chiral centers *without* a local plane of symmetry halfway between them.
- **Isotactic** (polymer): A polymer containing only meso diads.
- **Syndiotactic** (polymer): A polymer containing only racemic diads.
- **Atactic** (polymer): A polymer containing both meso and racemic diads.
- Example: Atactic polystyrene tends to be more amorphous than syndiotactic or isotactic polymers, which can be semicrystalline.
 - Nobel prize (1963) to Ziegler and Natta for a catalyst generating isotactic polystyrene (PS-it).
 - Note that the initial Ziegler-Natta catalysts weren't metallocenes! The introduction of these types only came later.
 - Syndiotactic polystyrene (PS-st) came later in 1986 and had superior properties.
 - PS-st crystallizes an order of magnitude faster than PS-it; has half the entanglement molecular weight; and is commonly used today in auto parts, electronics, and medical equipment.
- Tacticity is often measured by certain splittings (or their absence) in ^1H NMR.
 - Isotactic polymers put the geminal methylene protons into distinct chemical environments; syndiotactic polymers do not. Atactic polymers will have a mix of both, and the mix can be quantified with integration.
 - ^{13}C NMR can be used, too.
- More on geometric isomers.
 - Example: Polybutadiene can be formed *trans* or *cis*, as guided by a catalyst.
 - Natural rubber is *cis*-1,4-polyisoprene. Other types of tree sap can give *trans*-1,4-polyisoprene.
- We now move onto polymer conformations.
- **Conformations:** The spacial arrangements possible (or "probable," taking energy into account) for a polymer.
 - No bonds are *broken* here, just rotated.
- Reviews **Newman projections**.
 - Staggered configuration is defined as 0° .
 - Eclipsed configuration then starts at 60° .
 - There are energy penalties to being in different conformations.
 - As one example, eclipsed is higher energy than staggered due to sterics.
 - Generally sinusoidal relation in a plot of potential energy V against dihedral angle θ .
 - The energy difference between rotamers is approximately $3k_{\text{B}}T$, which is not huge but big enough that the system will spend most of its time in the valleys. Each "valley" is a **conformer**.
 - So then since probability is proportional to $e^{-V/k_{\text{B}}T}$, the probability that a molecule will be staggered is greater than that it will be eclipsed.
 - In molecules longer than ethane, we break degeneracy of the valleys.
 - The rapid growth of conformers: Ethane has 3 conformers. Propane has 3^2 . Butane has 3^3 . Decane has 3^{10} . Polyethylene with $N = 10^5$ already has on the order of 10^{47000} possible conformers, a huge conformational space.

- This is because each bond has 3 valleys!
 - Many of the models we'll develop are ways of enumerating these conformations in relation to some higher-order measurement of the polymer, such as the **n-band difference**.
 - Polymers are indeed often moderately sized coils rather than fully stretched out rods.
 - Example: 166 kbp DNA (approximately 684 **Kuhn steps**, discussed next lecture) can be videotaped moving around, and it never fully elongates.
 - Stretched out polymers shrink back over some characteristic time.
- Polymers with high degrees of polymerization result in many possible conformations without breaking bonds — this is what we'll discuss in the next two lectures!
- A good conversions to keep in mind: $2.5 \text{ kJ/mol} \approx 1k_B T$.
 - Today is probably the most jampacked bits and pieces day; other lectures will be more focused, but this is important background.

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