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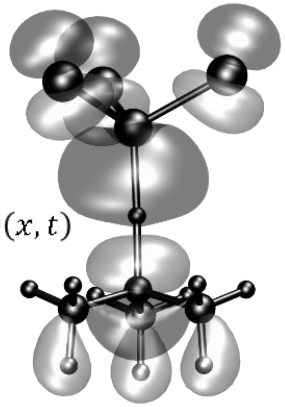
Computational Chemistry@SCAI.VMD

QM

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V(x,t)\right]\psi(x,t) = E\psi(x,t)$$



2nd order DE

- atomic positions, multiplicity, basis sets & level of theory

→ atomic/molecular properties: IP, EA, dipolemoment, FF constants,….

computational demand

MD/MC

$$E_{pot}(\vec{r}_1,\ldots,\vec{r}_n) = \sum_{bonds} k_r(r-r_0)^2 + \sum_{angles} k_\vartheta(\vartheta-\vartheta_0)^2 + \frac{1}{2}\sum_{dihedrals} V_n(1+\cos(n\varphi-\gamma)) + \sum_{nonb}\left(\frac{A}{r^{12}} - \frac{C}{r^6} + \frac{q_i\cdot q_2}{D\cdot r}\right)^2$$

system of 2nd order DEs/ Markov chain

- atomic positions, FF, p,T

→ physical (bulk) properties: density, VLE, viscosity, conductivity, surface tension, …

computational demand, no reactions

kinetics

$$\frac{d[P]}{dt} = k\cdot[A][B]$$

system of ODEs

- reactions, rate constants, concentrations,p,T

→ macroscopic (dynamic) behaviour: concentrations, thermic/pressure effects,..

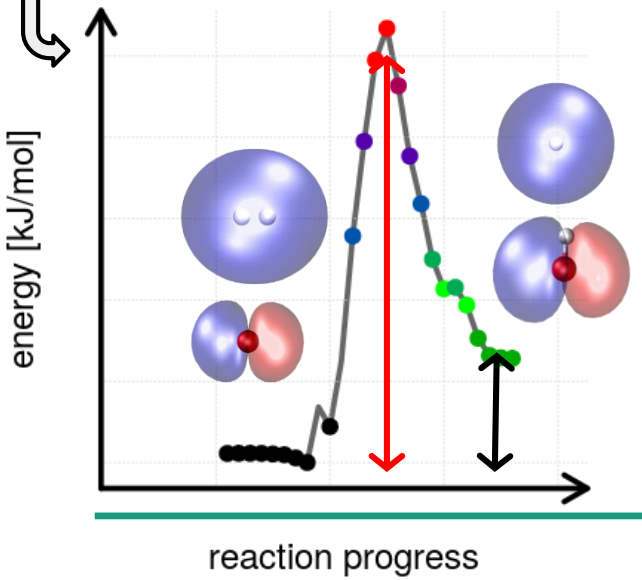
Fraunhofer SCAI

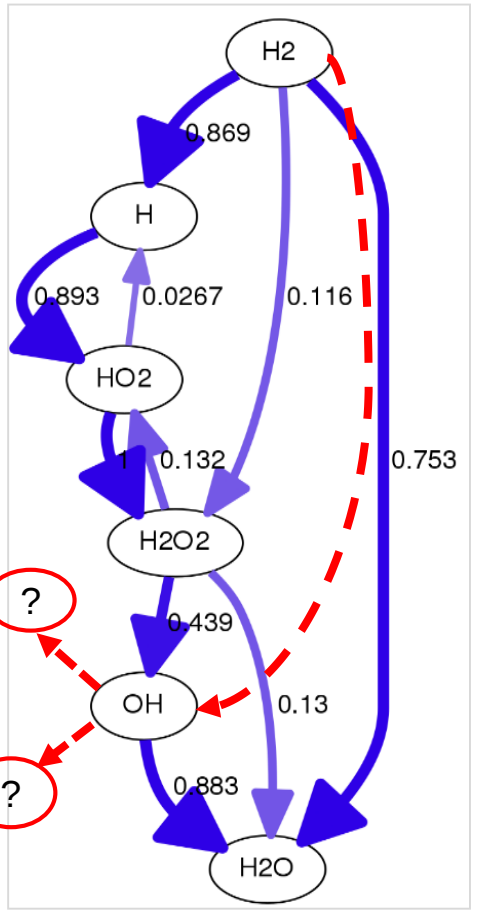
oxyhydrogen reaction

$$2\,H_2 + O_2 \rightarrow 2\,H_2O$$

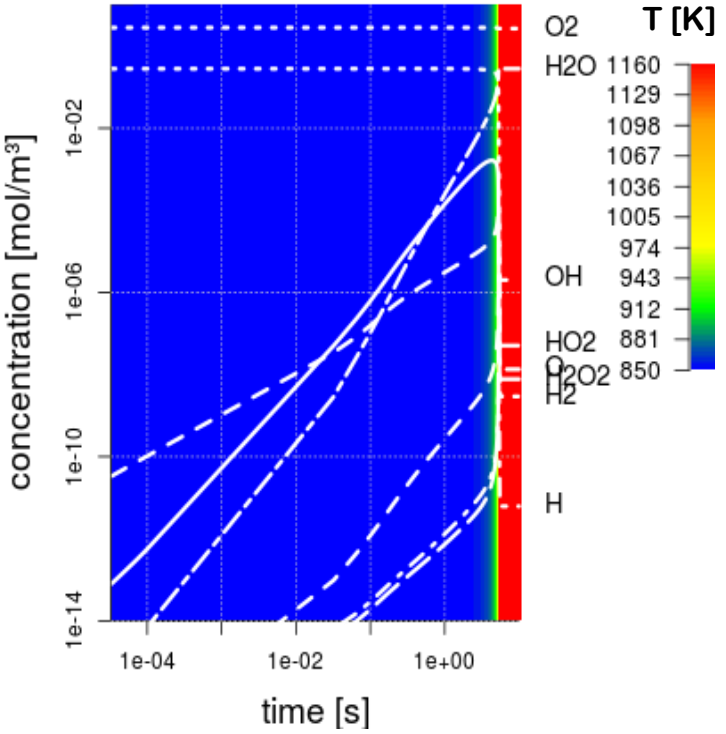
reactants	products	E barrier	E gain/ E loss
H2	H + H	435	435
O2	O + O	495	495
O + H2	OH + H	42	6
....	....	....	....

$$A \cdot T^{p} \cdot e^{-\frac{E_b}{RT}} = k$$





reaction kinetics



system of ODEs

$$\frac{d[p]}{dt} = k \cdot [r]^{n}$$

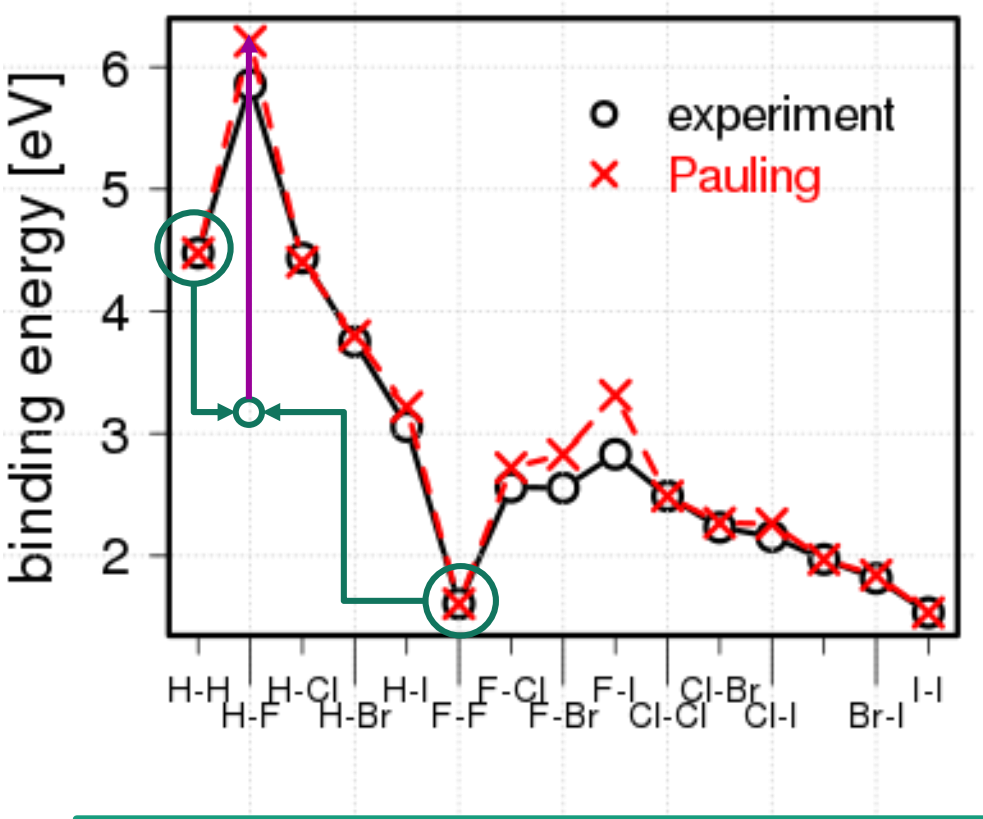
→ anticipate nodes
→ anticipate edges

→ **learn molecular energies from atomic features**

- rapid
- exothermic
- production of H_2O

Fraunhofer SCAI

predicting energies for A—B-type molecules



Pauling 1932

- introduced *electronegativity*
- reproduced trends by:

$$E_{AB} = \frac{E_{AA} + E_{BB}}{2} + (EN_A - EN_B)^2$$

→ find formula for E_{AB} by modern learning techniques

Fraunhofer SCAI

GLMNET

- choose energy related atomic properties, IP, EA, EN, E_{AA}
- compile **input matrix**
  - primary features
  - sums and differences
  - products of 1 and 2

  → > 350 entries
- **binding energies**
- solve

$$\min_{\beta_o,\beta} \frac{1}{N}\sum_{i=1}^{N} w_i l(y_i, \beta_o + \beta^T x_i) + \lambda\left[\frac{(1-a)\|\beta\|_2^2}{2} + \alpha\|\beta\|_1\right]$$

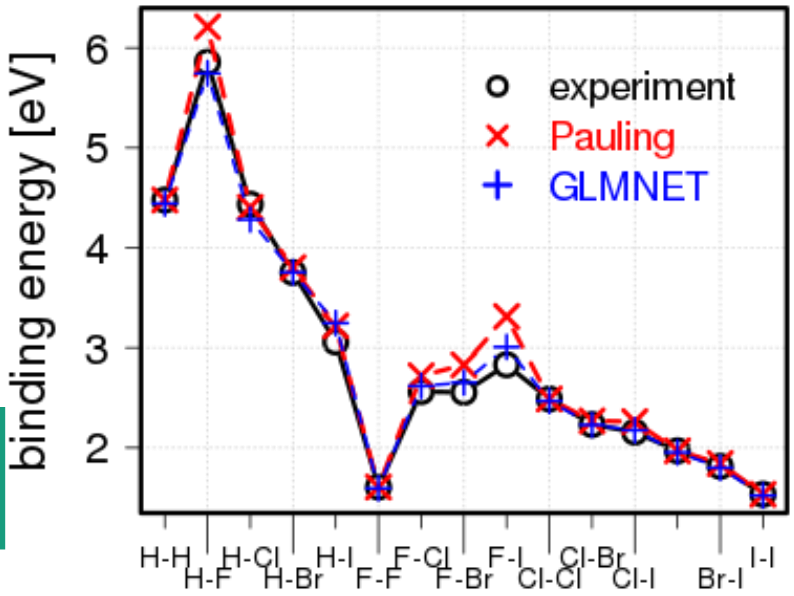
A	B	IP_A	IP_B	...	$\Delta IP \cdot EN_A$	y
H	H	13,6	13,6	...		4,48
H	F	13,6	17,6	...		5,86
H	Cl	13,6	13,0	...		4,44
H	Br	13,6	11,9	...		3,75
H	I	13,6	10,6	...		3,06
F	F	17,6	17,6	...		1,61
...	...	...	...	...		...
I	I	10,6	10,6	...		1,53

→ most descriptive properties identified

→ proceduce robust to setup of property matrix

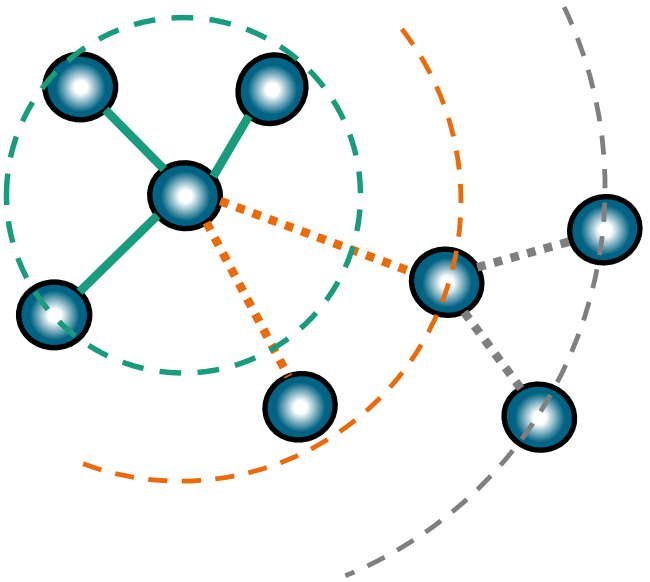
$$E_{AB} = \frac{0.998 \cdot (E_{AA} + E_{BB})}{2} + 0.85 \cdot (EN_A - EN_B)^2$$

Pauling: $$E_{AB} = \frac{E_{AA} + E_{BB}}{2} + (EN_A - EN_B)^2$$



→ physics recovered reasonably well

Next steps



- proceed to molecules in general
- using accessible atomic properties
- deep convolution networks

  → bond-order like/reactive potentials

  for use in MD simulations →→ reactions in condensed phase

Thank you!