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Quasi-chemical theory for Ba^{2+} and Sr^{2+} hydration

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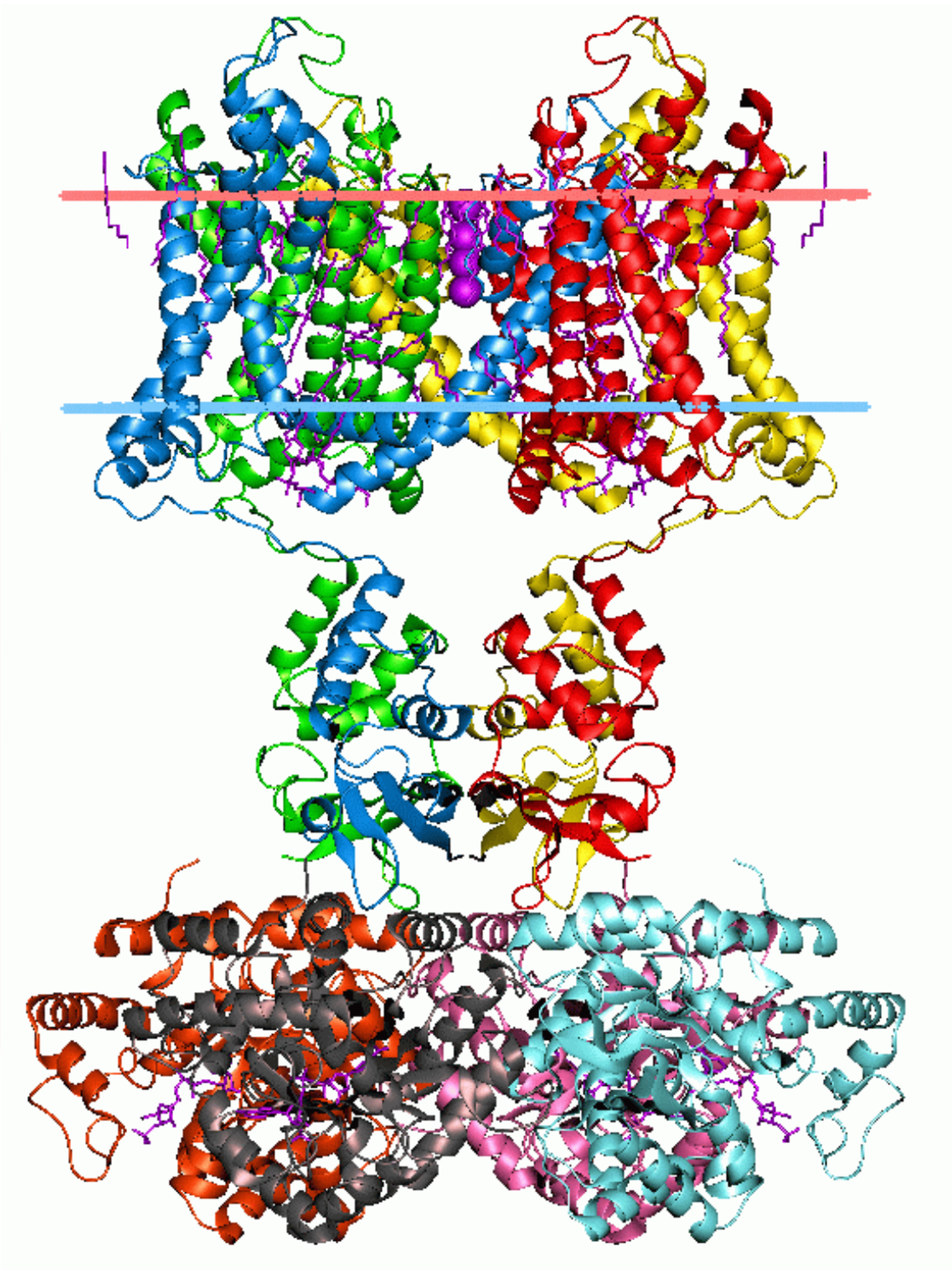
Telluride workshop: Ions in aqueous solutions and molecular biology, theory, simulation and modeling
July 8, 2014

Ion selective channels

- Ion selective channels
- Voltage gates
- Blockers barium



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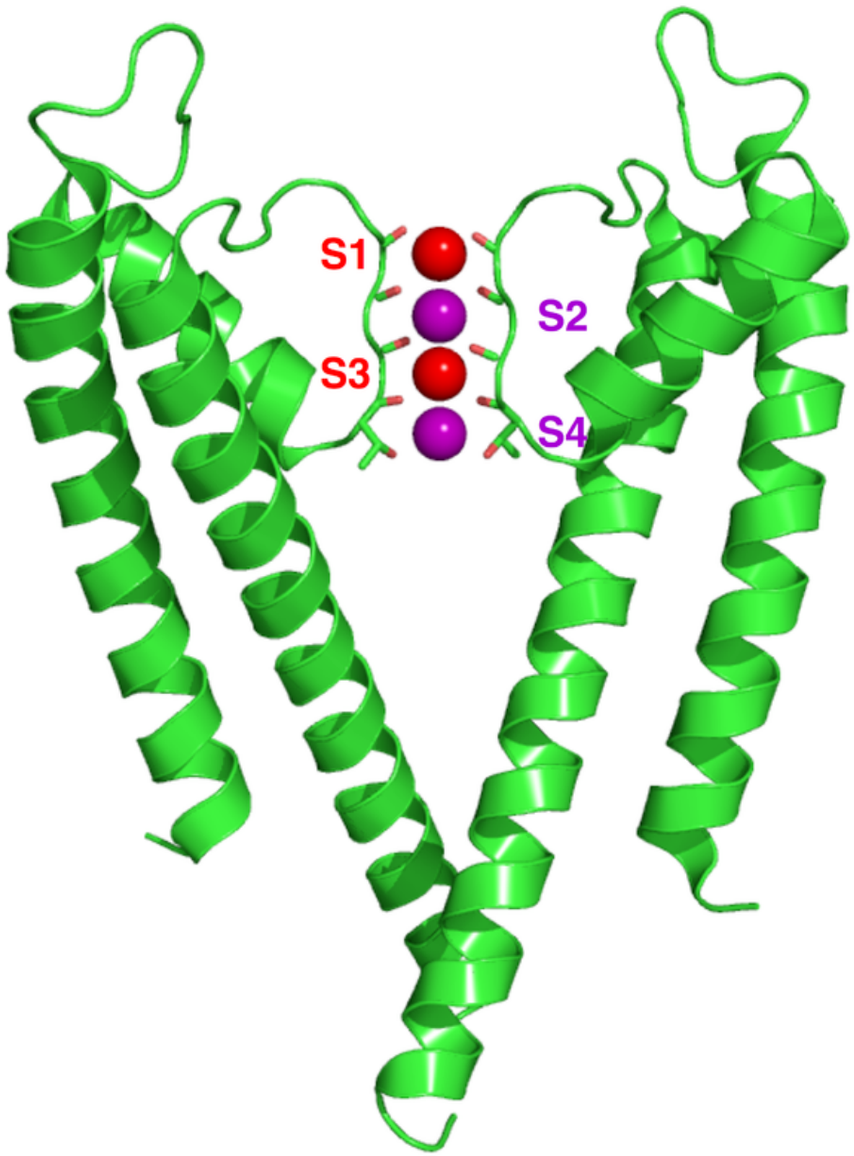
Barium blocking mechanism and controversies

- Where does Ba stick?
- Refer few papers Roux and Piasta miller etc



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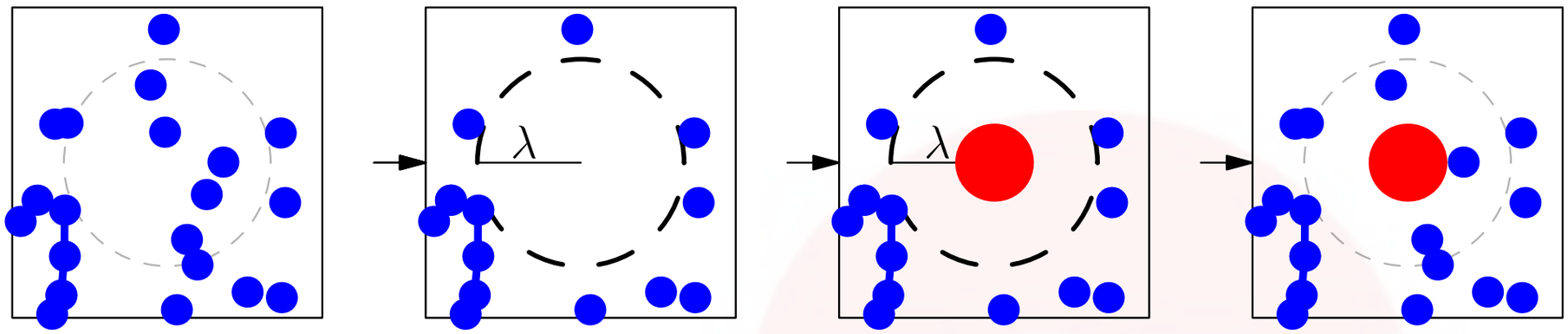
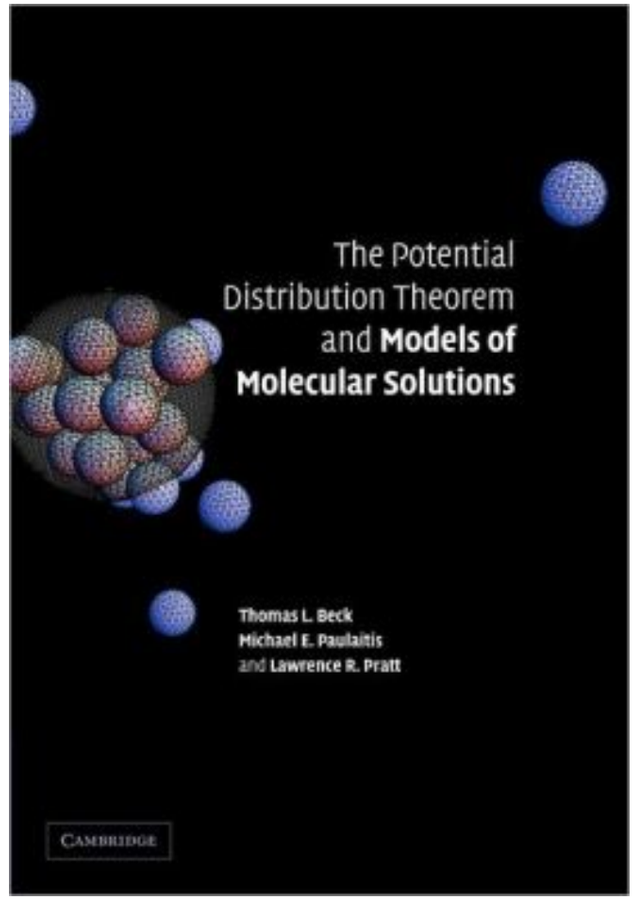
Quasi-chemical theory

$$\beta\mu_{\mathrm{w}}^{(\mathrm{ex})}\left(\varphi,p,T\right)=-\ln p^{(0)}(n_\lambda=0)+\ln\langle e^{\beta\varepsilon}\mid n_\lambda=0\rangle+\ln p(n_\lambda=0)$$





$$\ln\langle e^{\beta\epsilon}\mid n_\lambda=0\rangle=\beta\langle\epsilon\mid n_\lambda=0\rangle+\beta^2\langle\delta\epsilon^2\mid n_\lambda=0\rangle/2$$

- Packing
- Outer-shell
- Chemical

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QCT for ion hydration

$$\mu^{(\mathrm{ex})}_{\mathrm{Ba^{2+}}} = -kT\ln K^{(0)}_n \rho^n_{\mathrm{H_2O}} + kT\ln p_{\mathrm{Ba^{2+}}}(n) + \mu^{(\mathrm{ex})}_{\mathrm{Ba(H_2O)^{2+}_n}} - n\mu^{(\mathrm{ex})}_{\mathrm{H_2O}}$$

$$\mathrm{Ba^{2+}} + n\mathrm{H_2O} \rightleftharpoons \mathrm{Ba(H_2O)^{2+}_n}$$

Structural Models and Molecular Thermodynamics of Hydration of Ions and Small Molecules

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REVIEW

Case study of Rb^+(aq), quasi-chemical theory of ion hydration, and the *no split occupancies rule*

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Quasi-chemical study of Be^{2+}(aq) speciation

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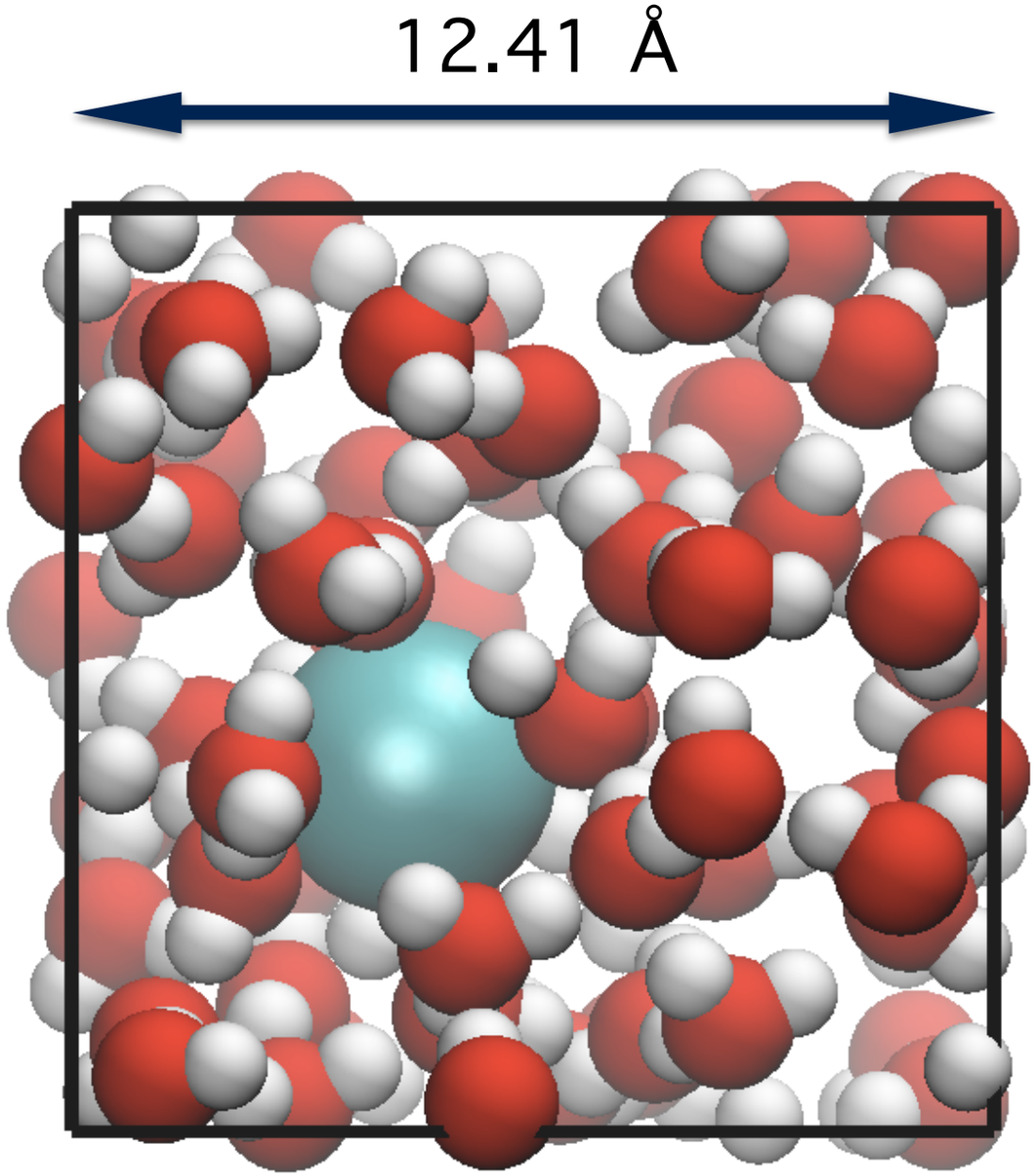
Barium

- Ab-initio MD (1 Ba^{2+}/64 H_2O)
- Electronic structure calculations
- Cluster approach
- Lambda selection



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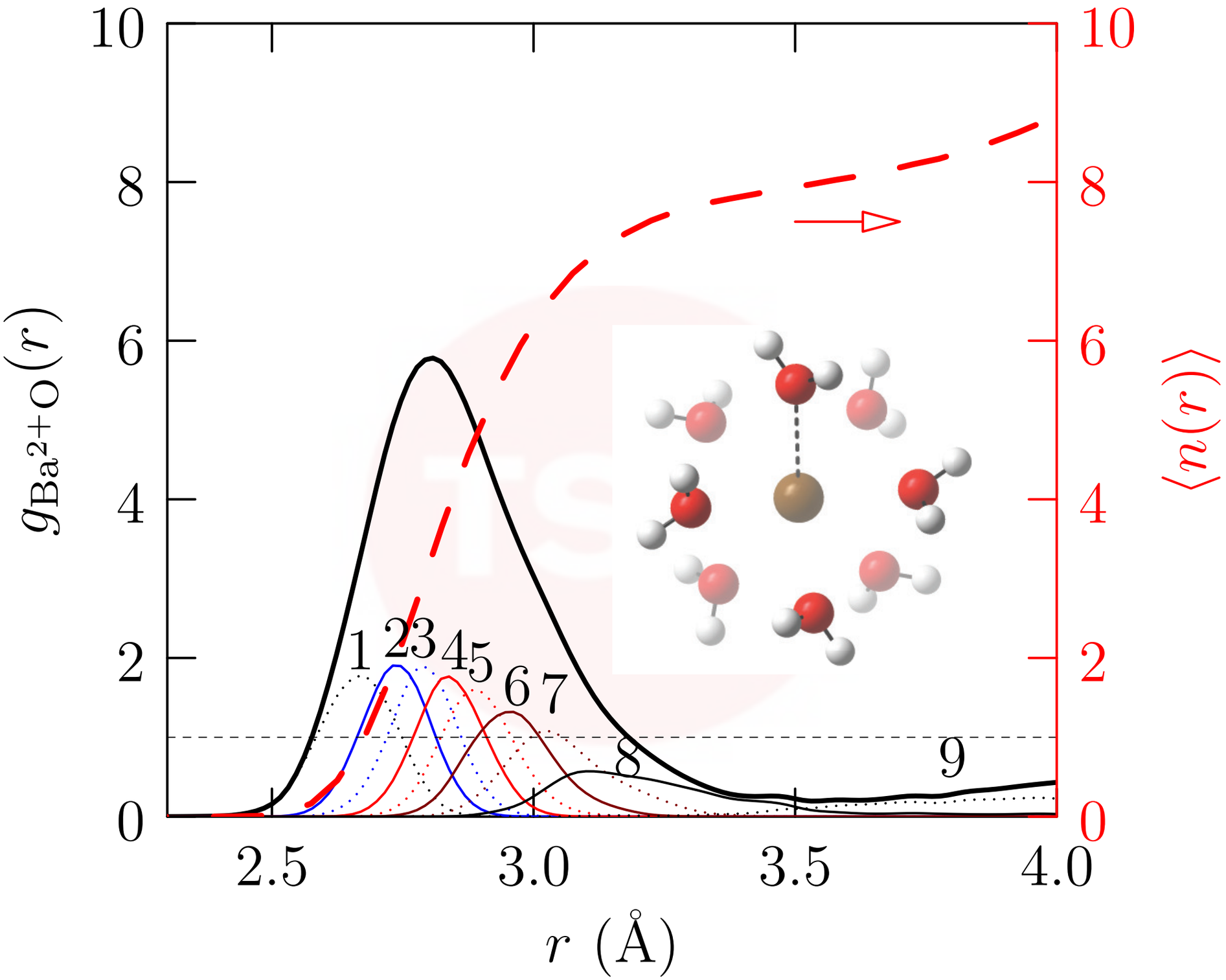
Structure of first inner shell



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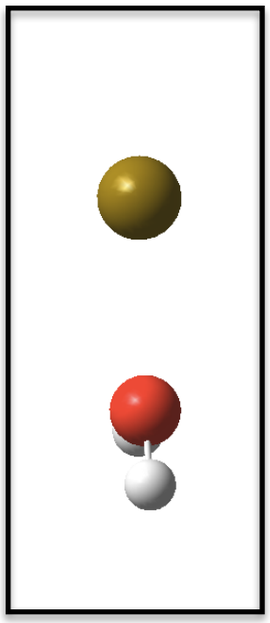
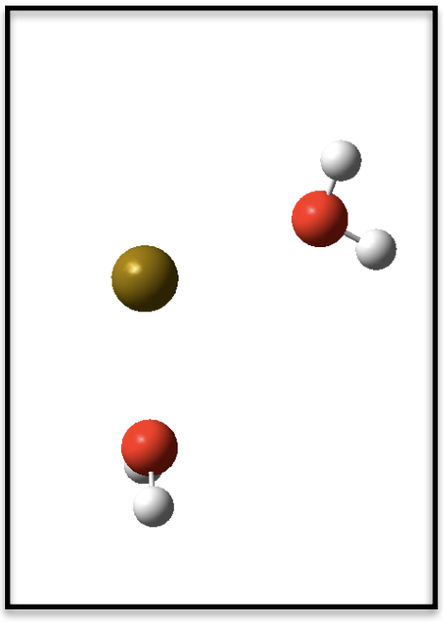
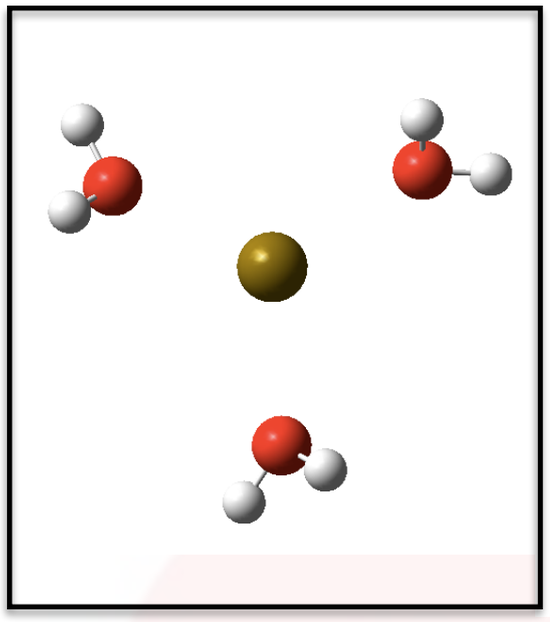
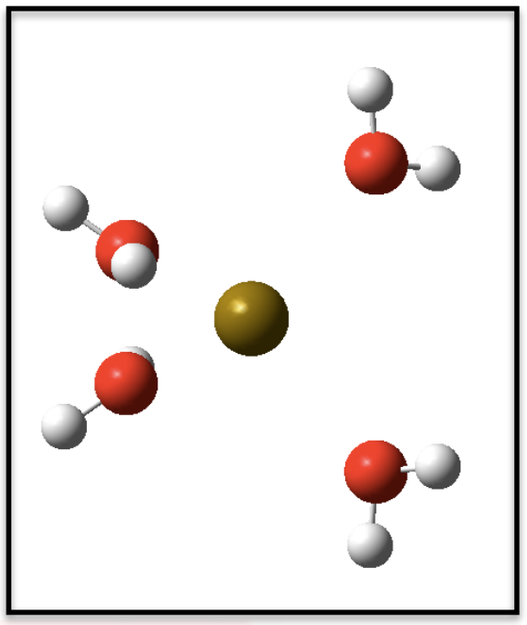
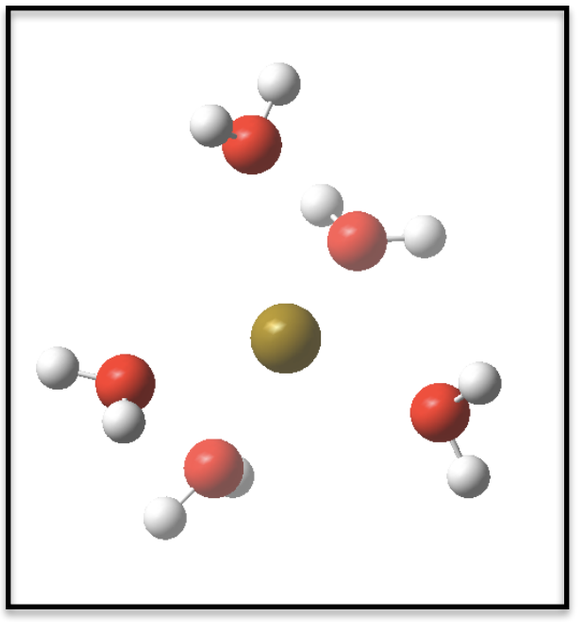
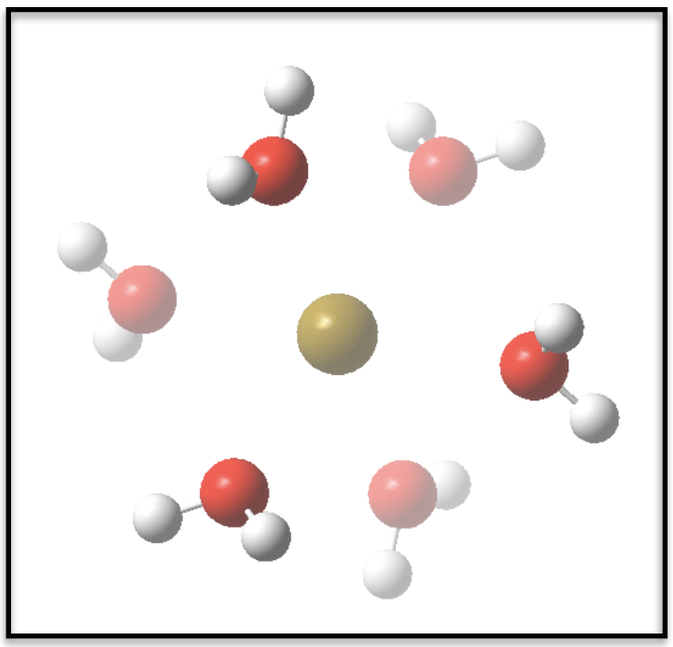
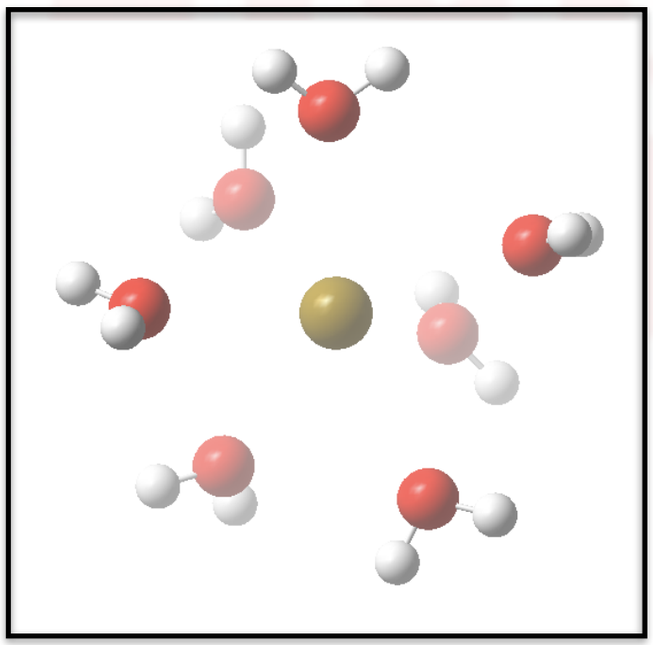
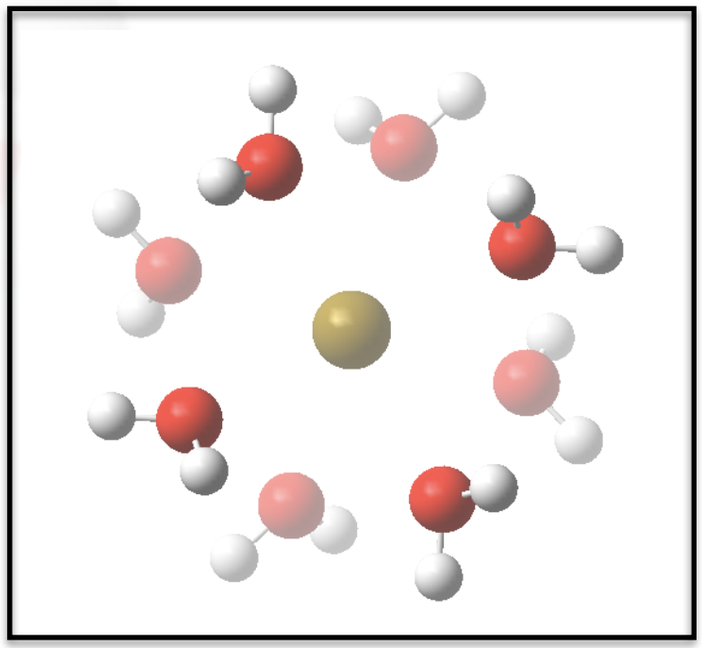
$Ba^{2+}(H_2O)_n$ clusters

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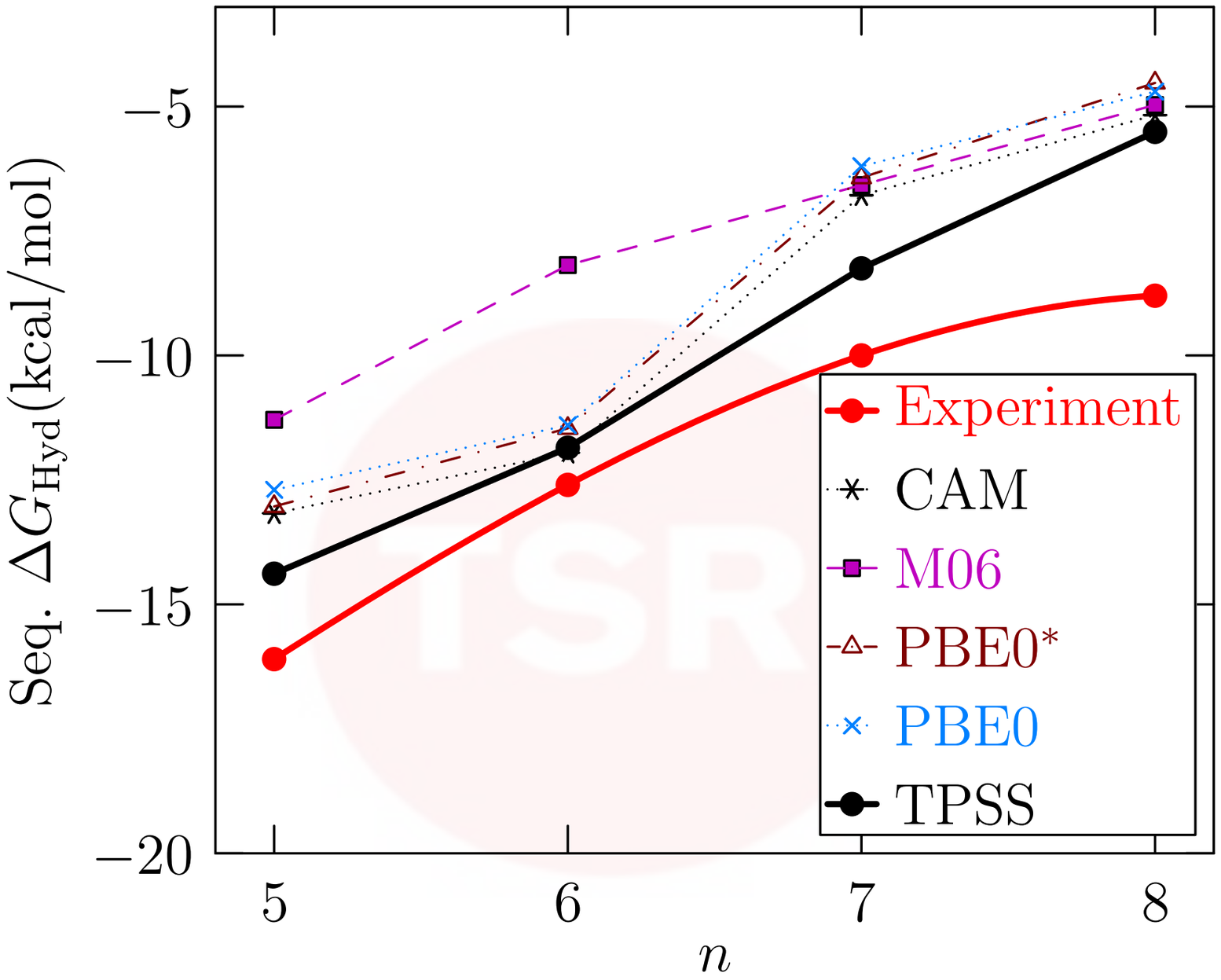
Selection of Functionals



experimental data from Peschke Kebarle paper, ref

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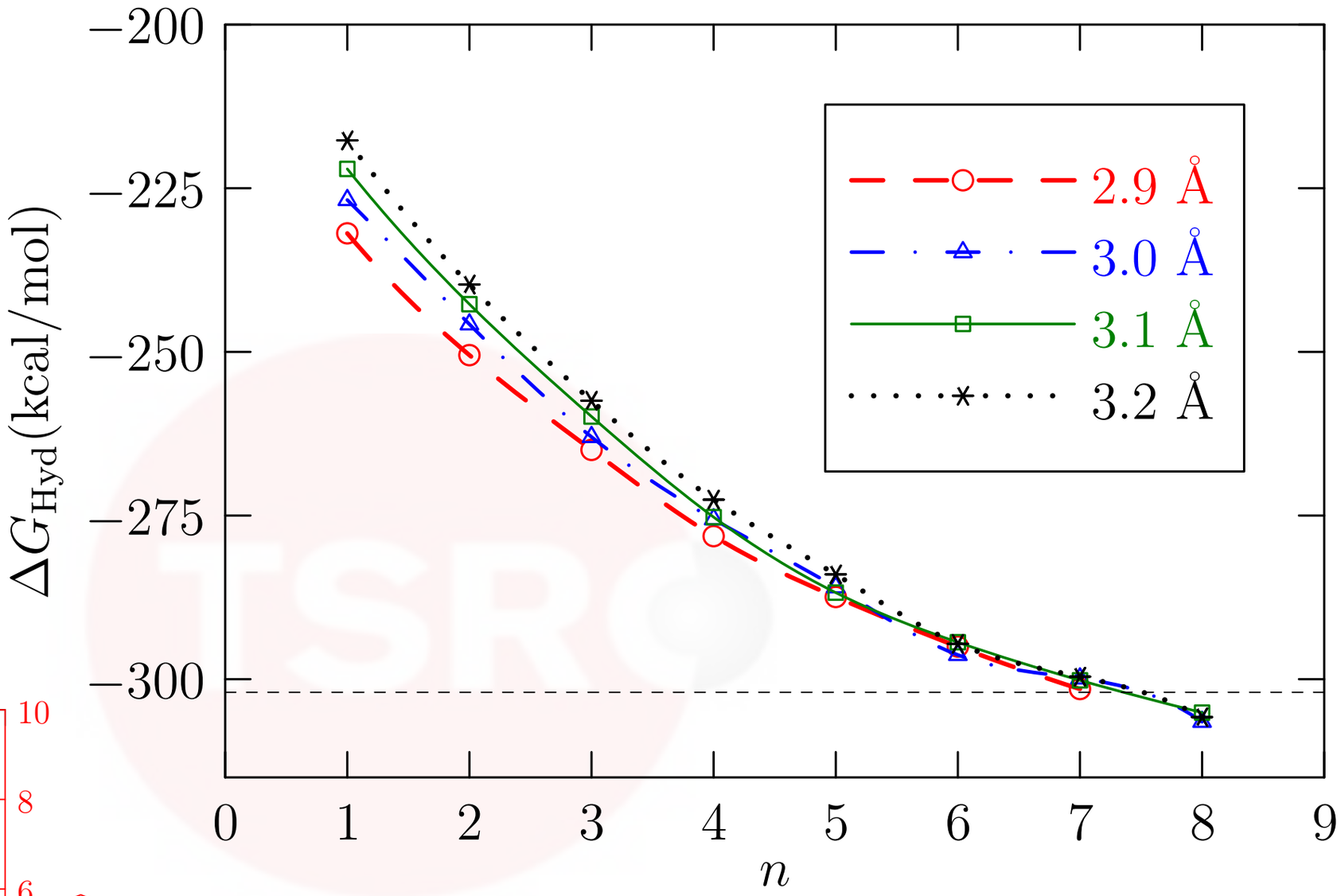
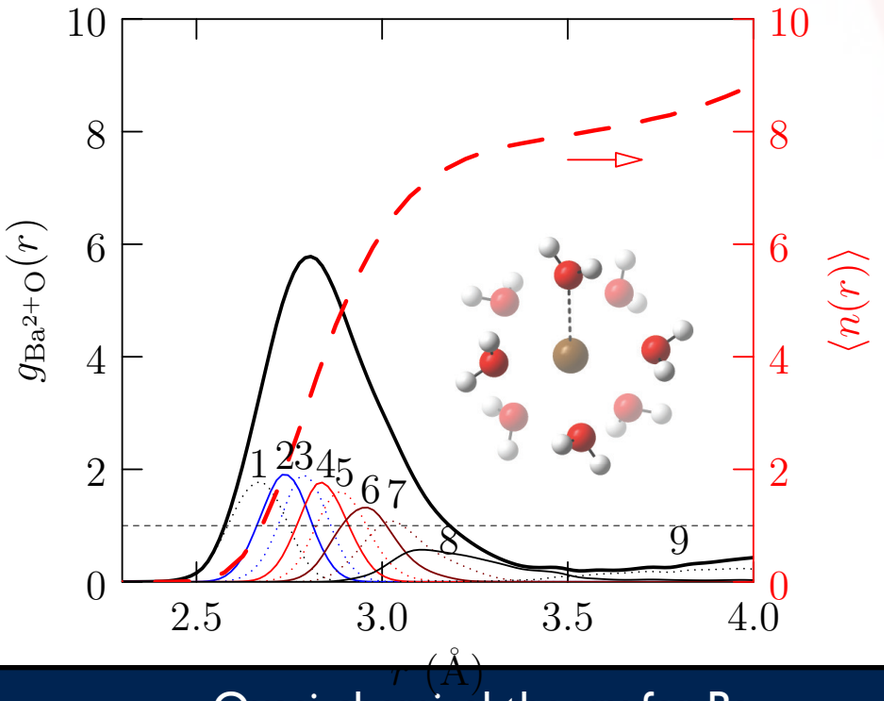
Hydration free energy





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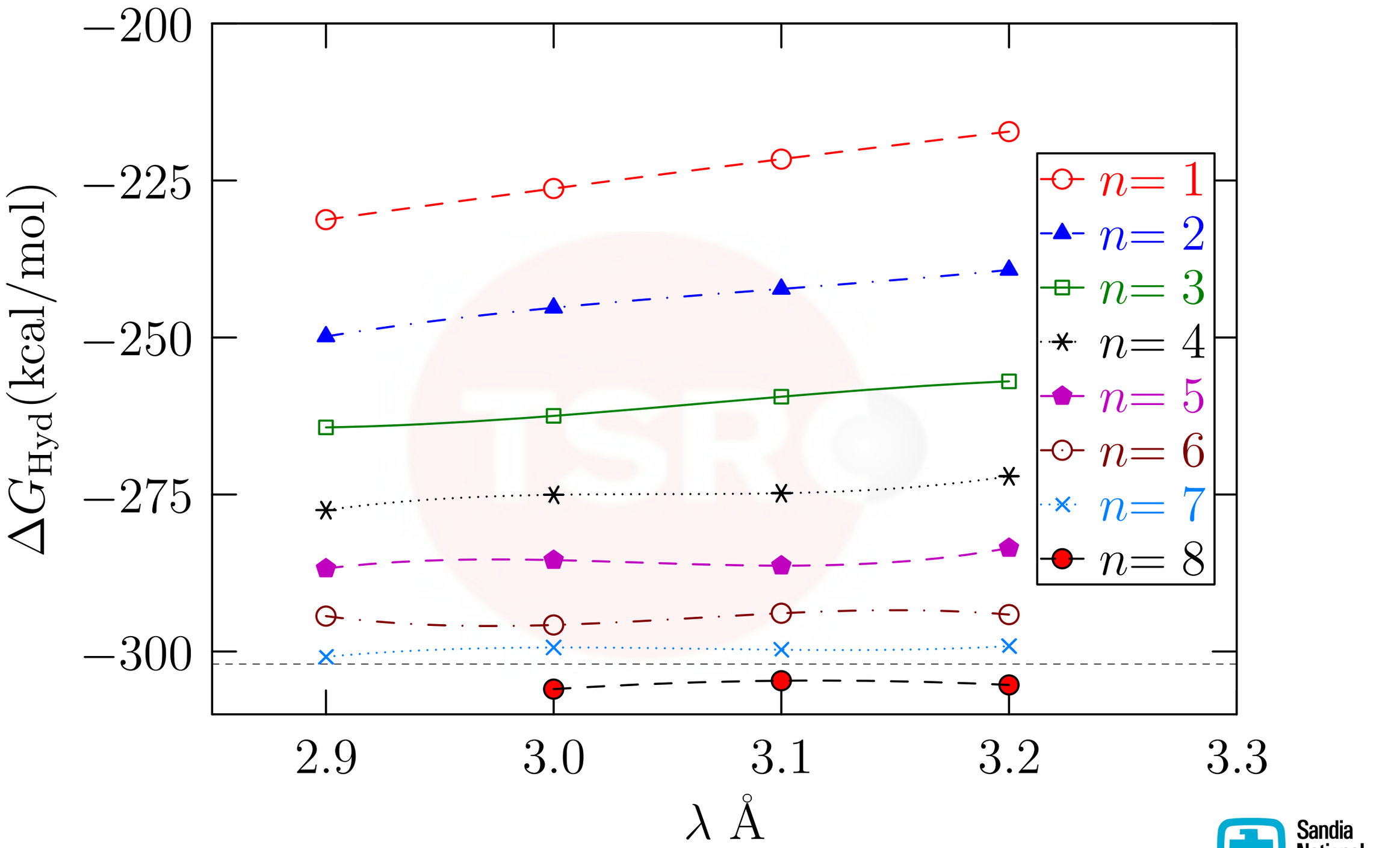
Hydration free energy



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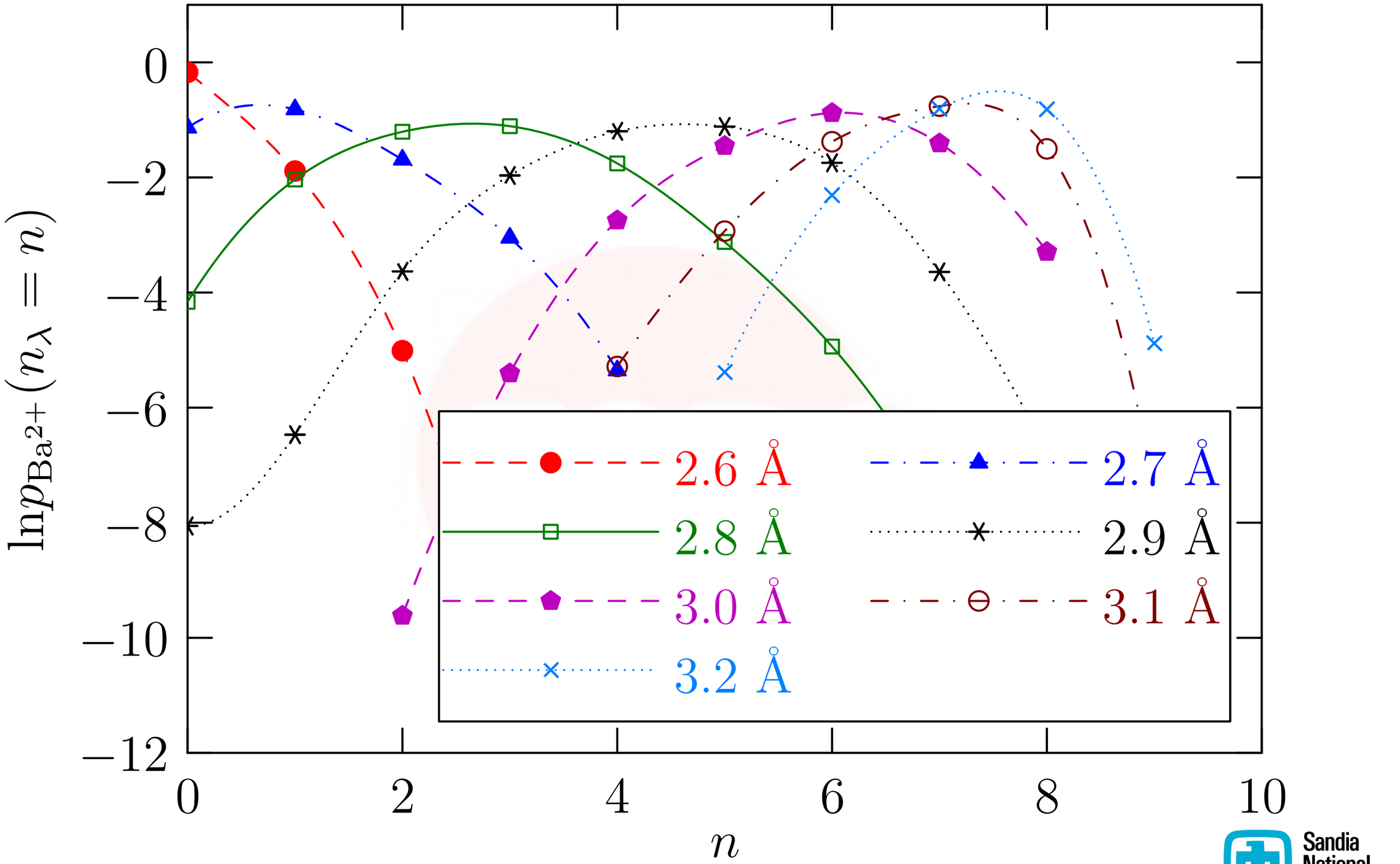
Occupancies of various inner shell definitions



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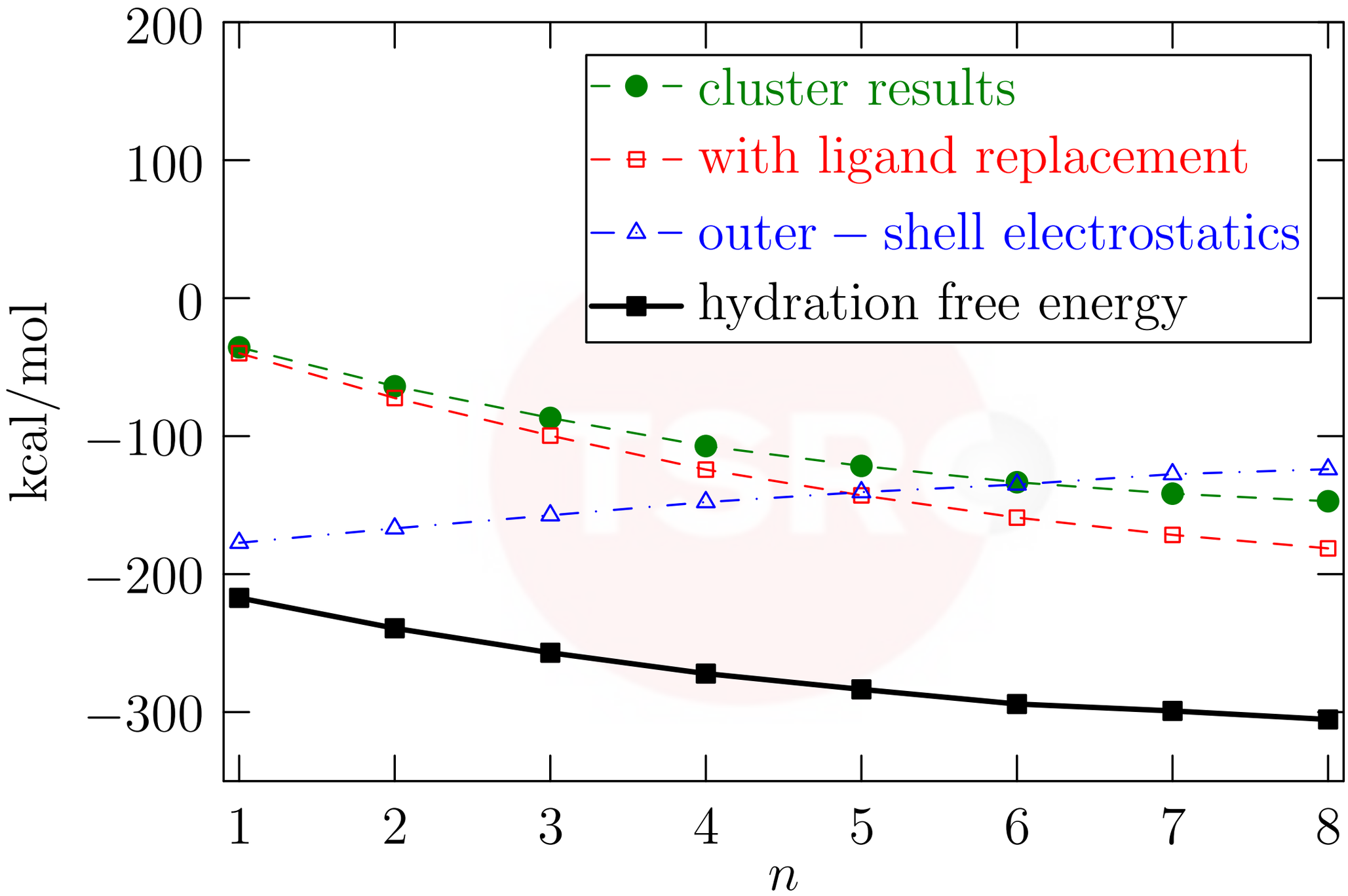
Different parts of hydration free energy



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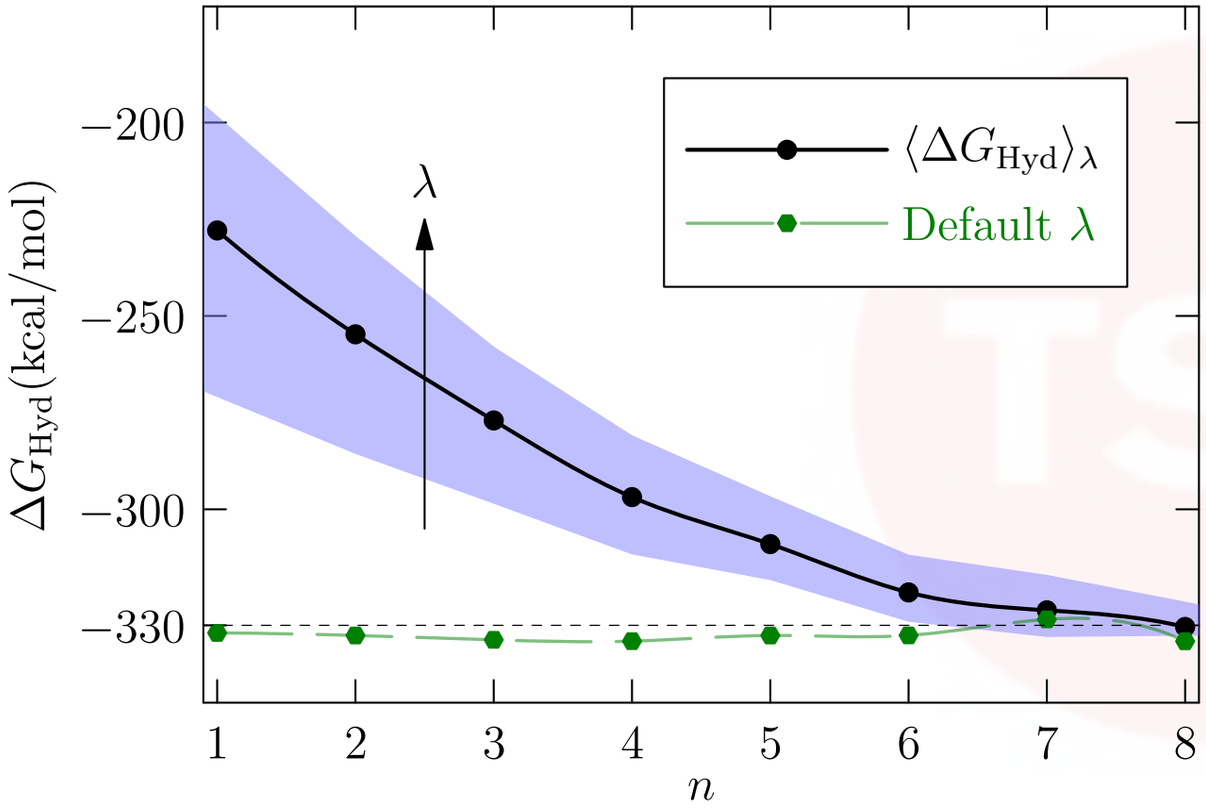
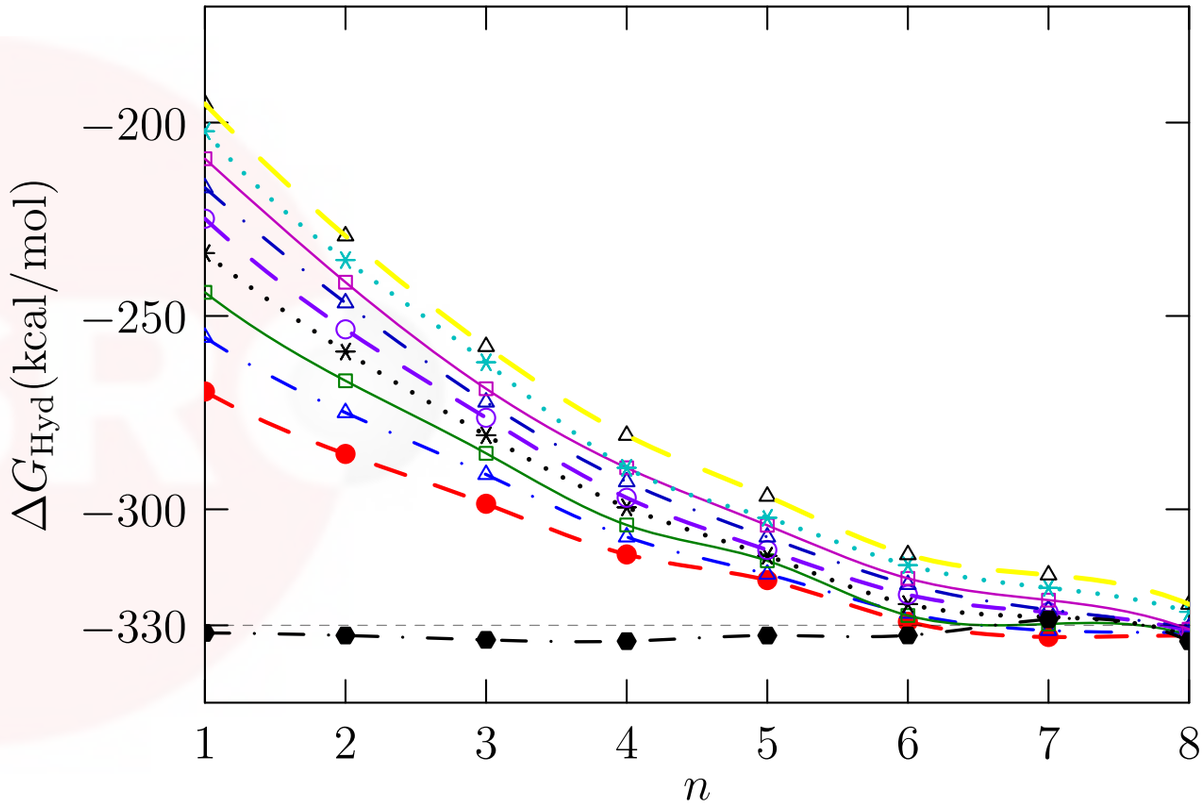
Strontium





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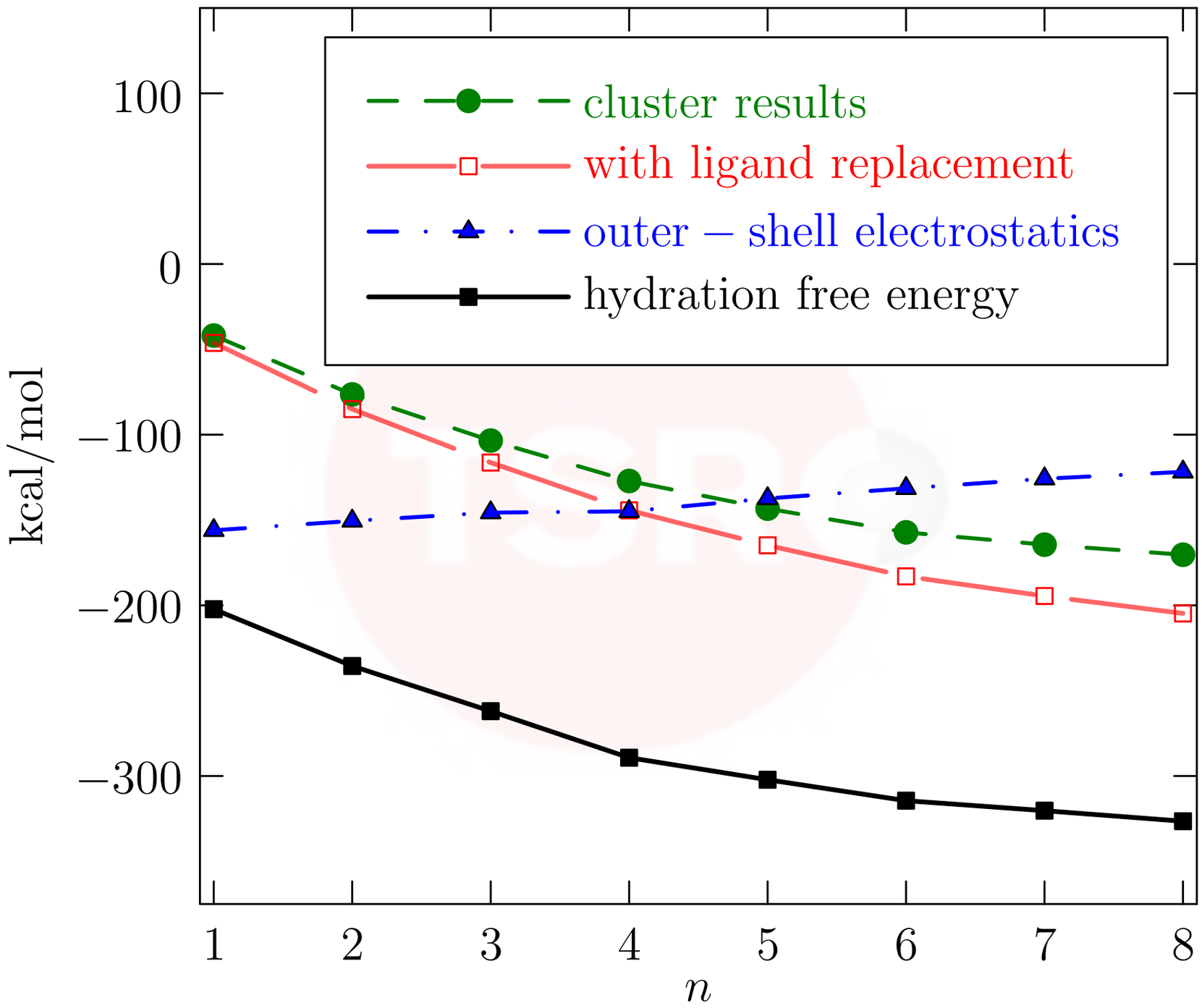
Hydration free energy



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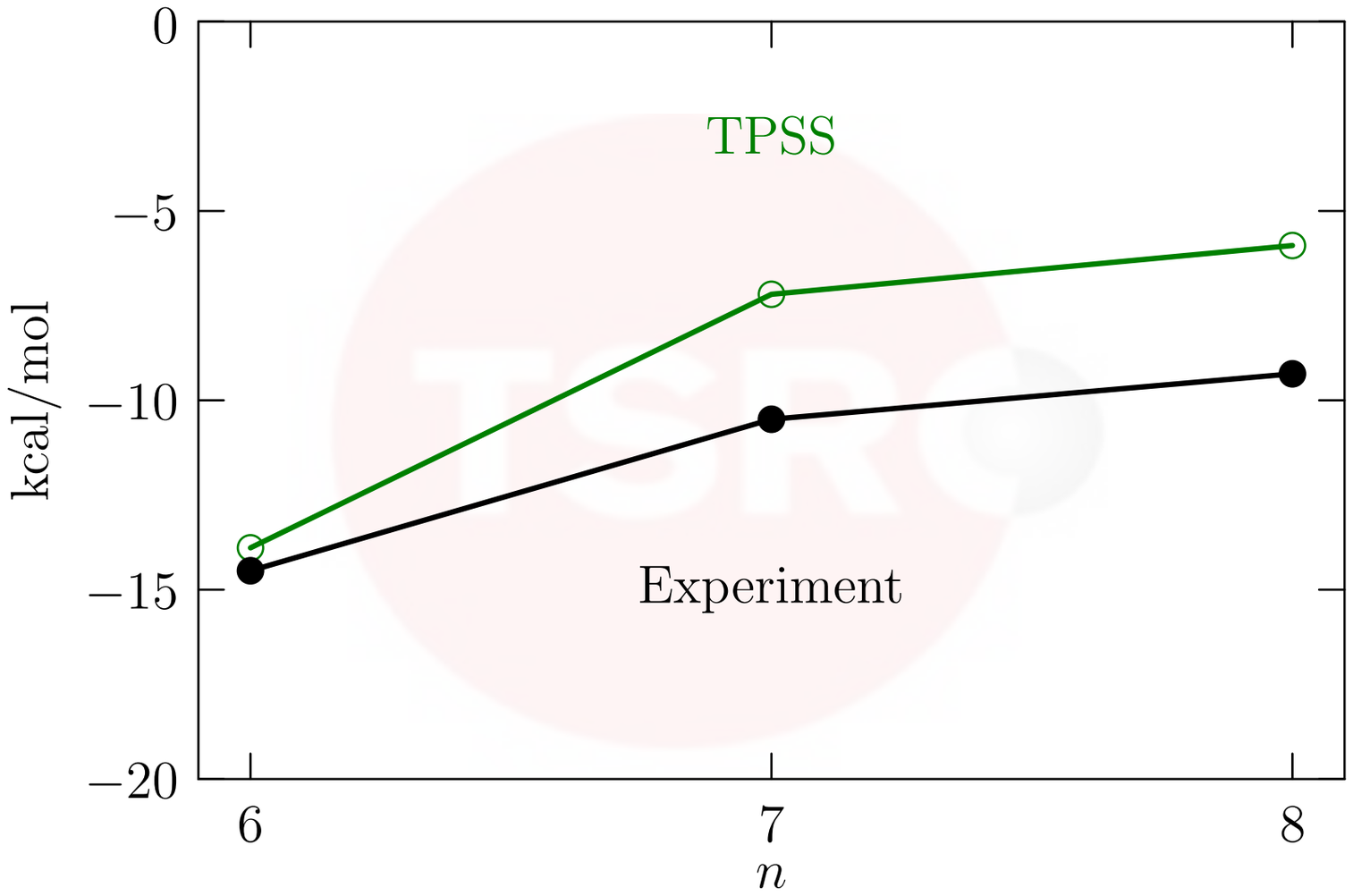
Sequential gas phase hydration free energy Strontium ion



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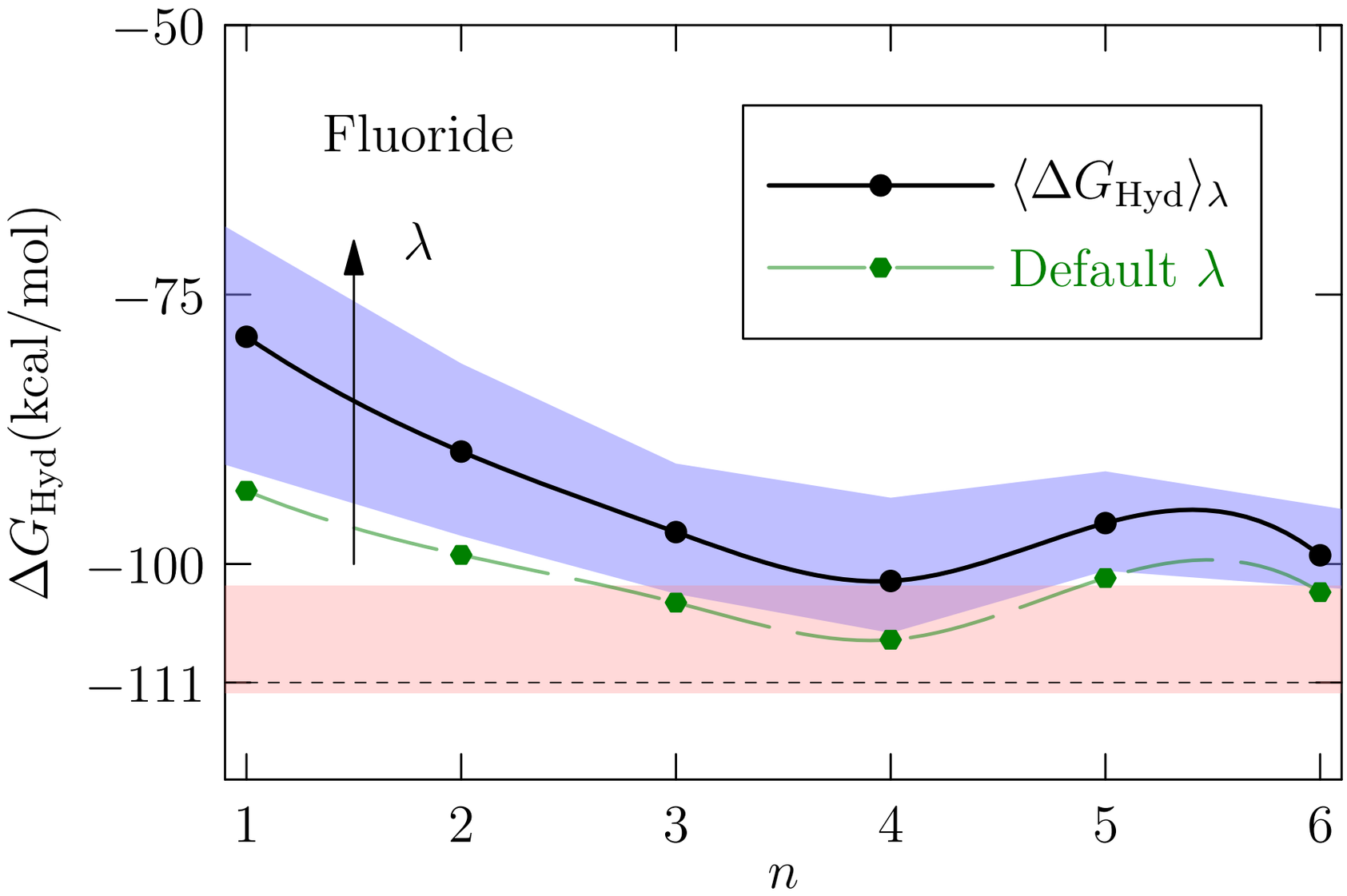
Fluoride



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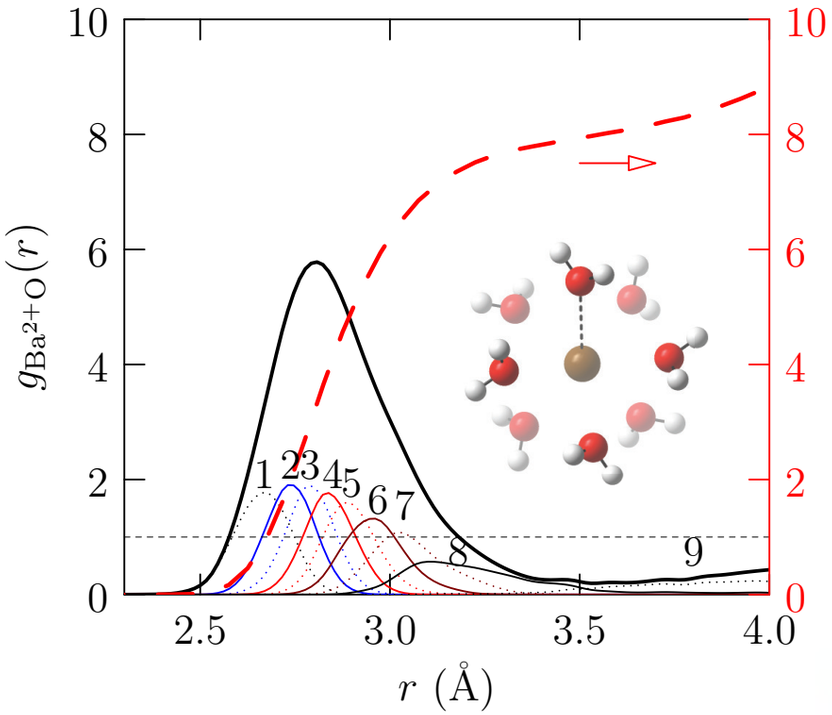
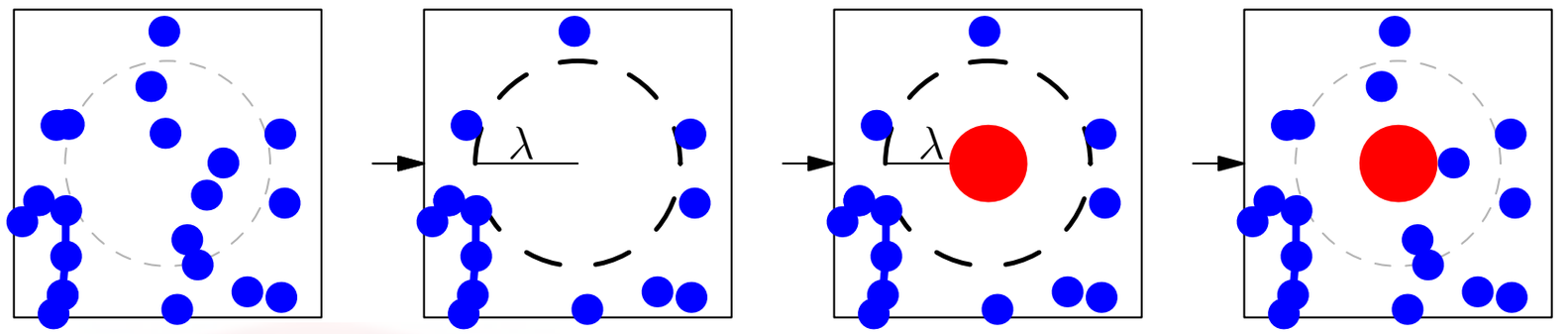
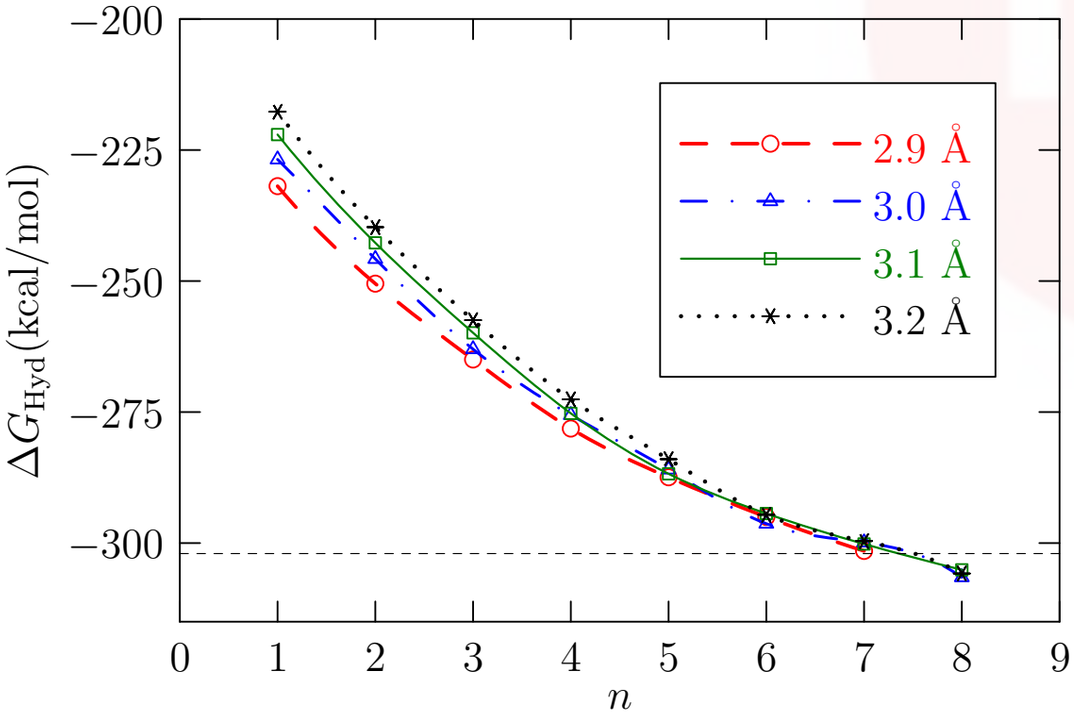
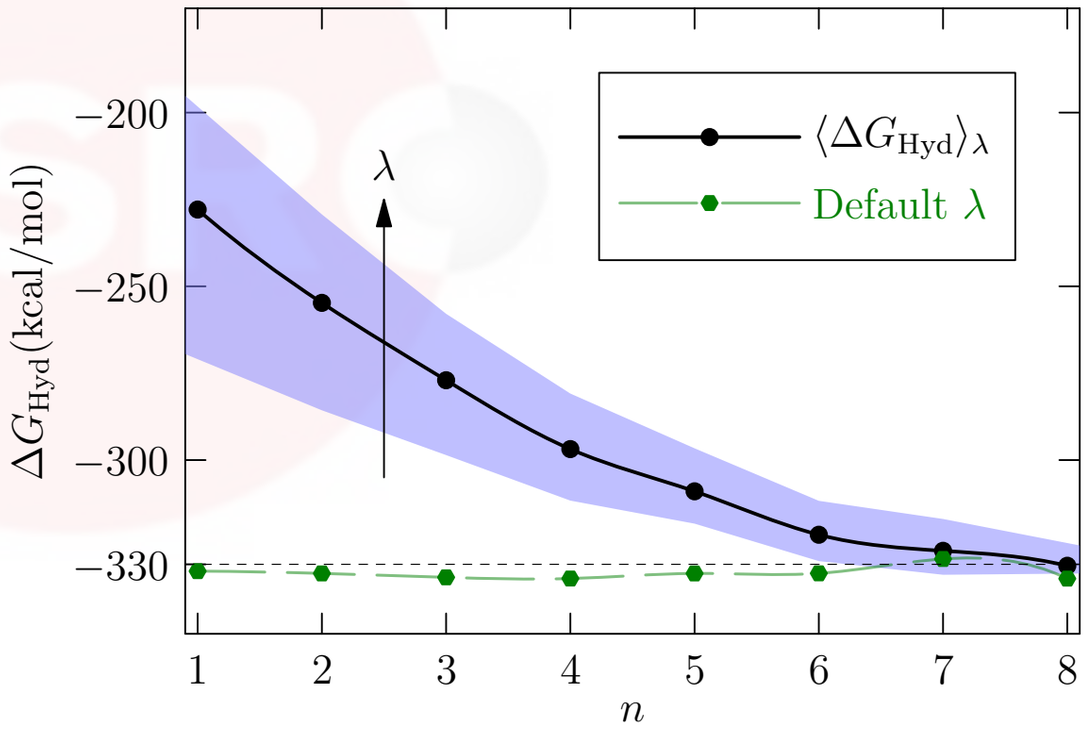
Summary









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