Faster G_0W_0 implementation for more accurate material design

By Jonathan Laflamme Janssen
Département de physique, Université de Montréal

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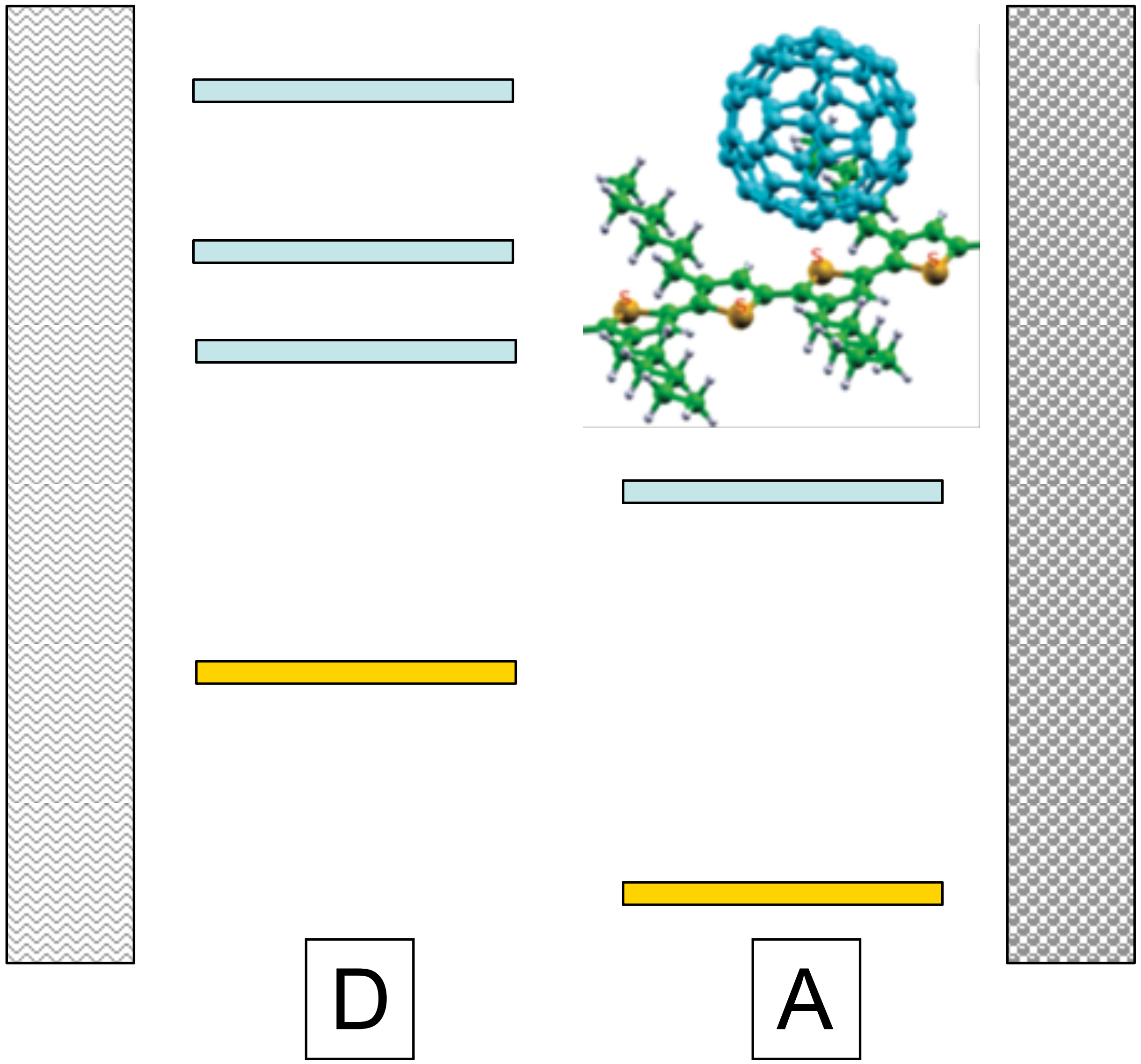
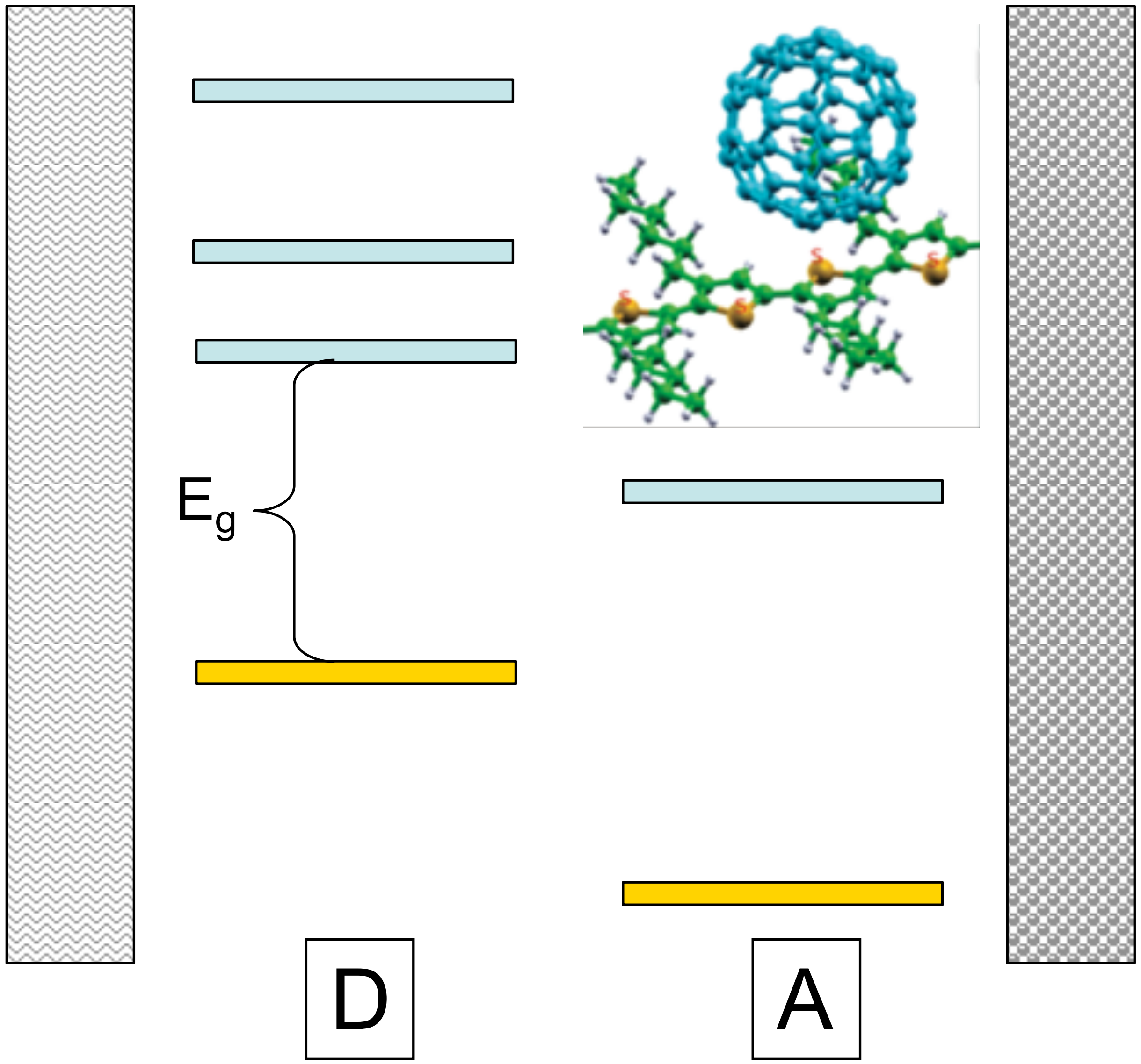
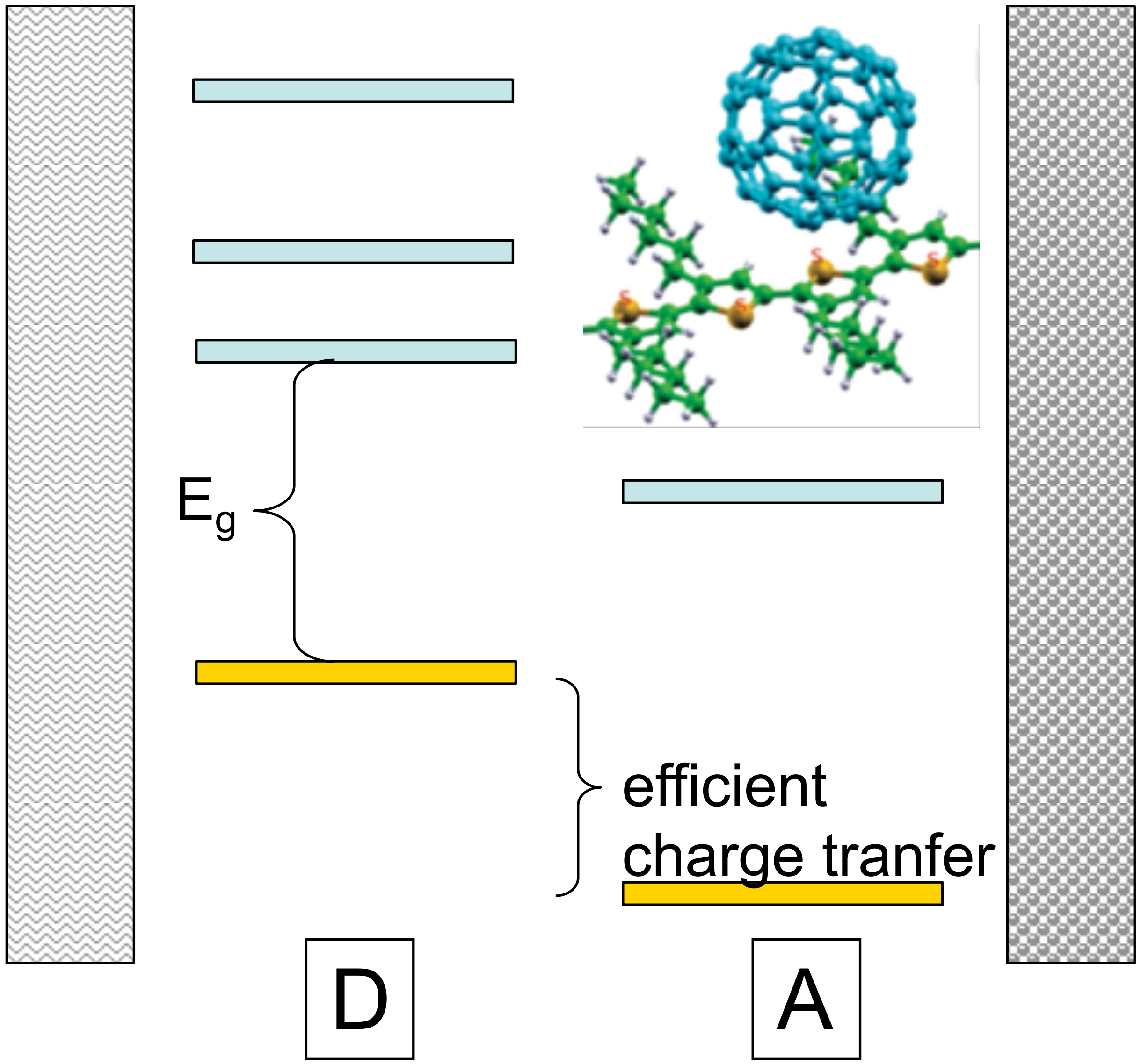
Design of polymers





- **Gap:** optimal for solar spectrum



- Gap: optimal for solar spectrum
- HOMO :
  - higher than C_{60}

    (for good charge transfer)

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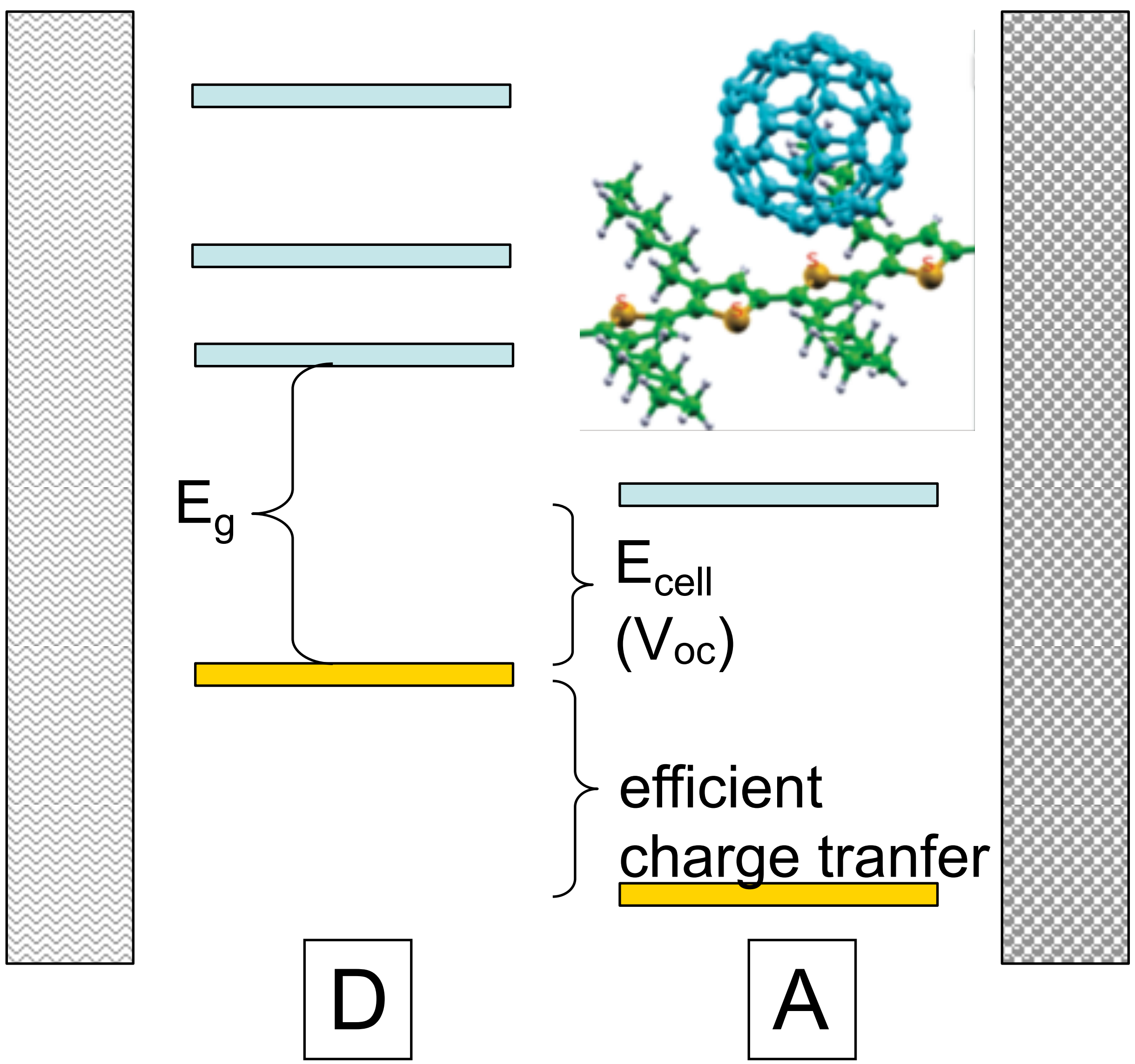

- Gap: optimal for solar spectrum
- HOMO :
  - higher than C_{60}

    (for good charge transfer)
  - low enough for good Voc

    and air stability

DFT to the rescue?

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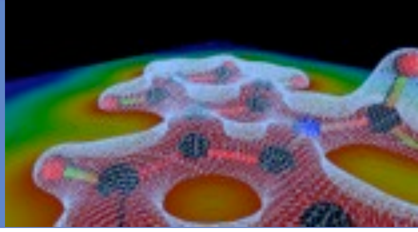

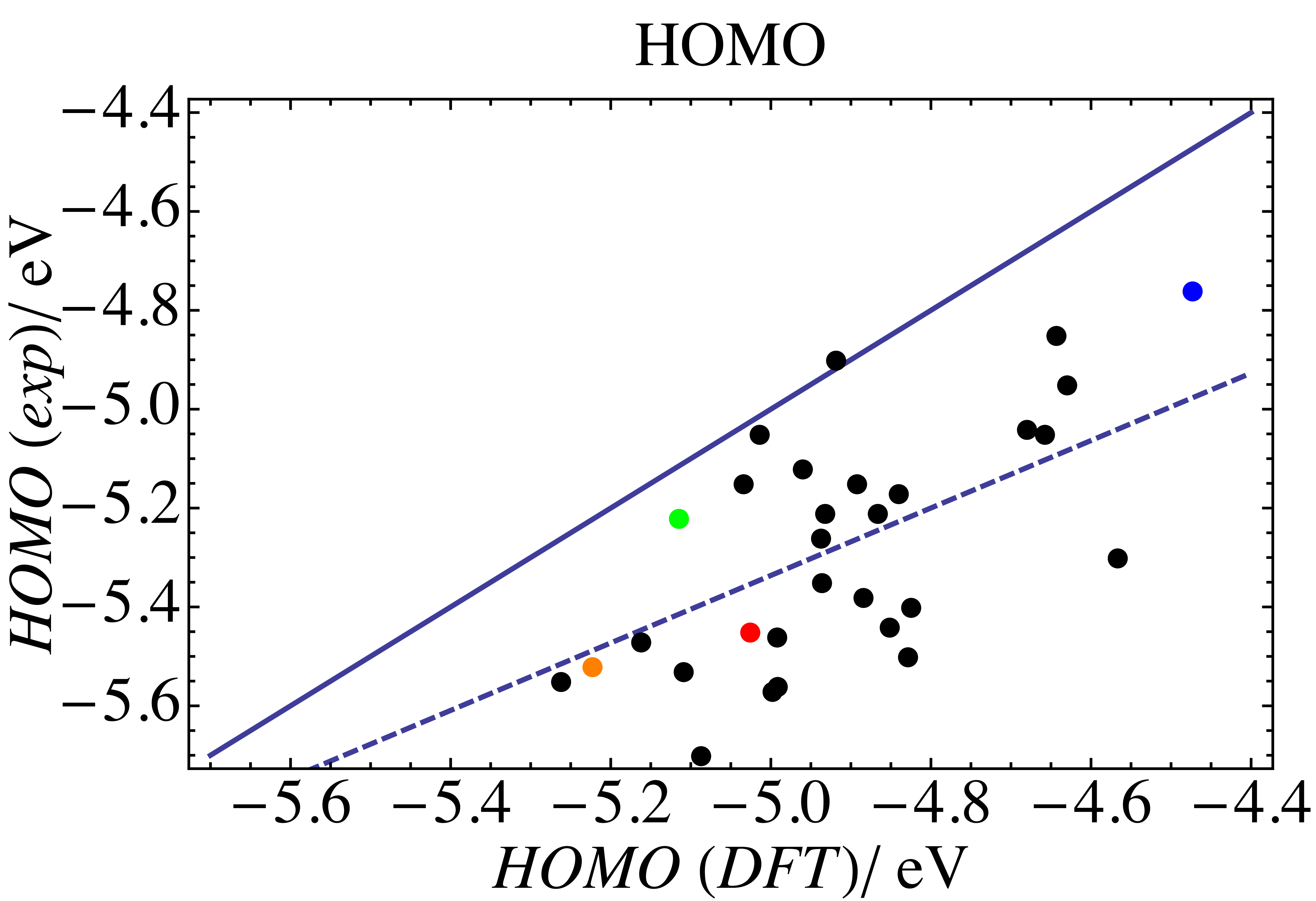

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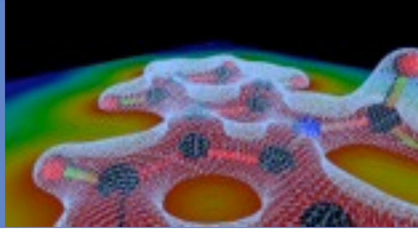
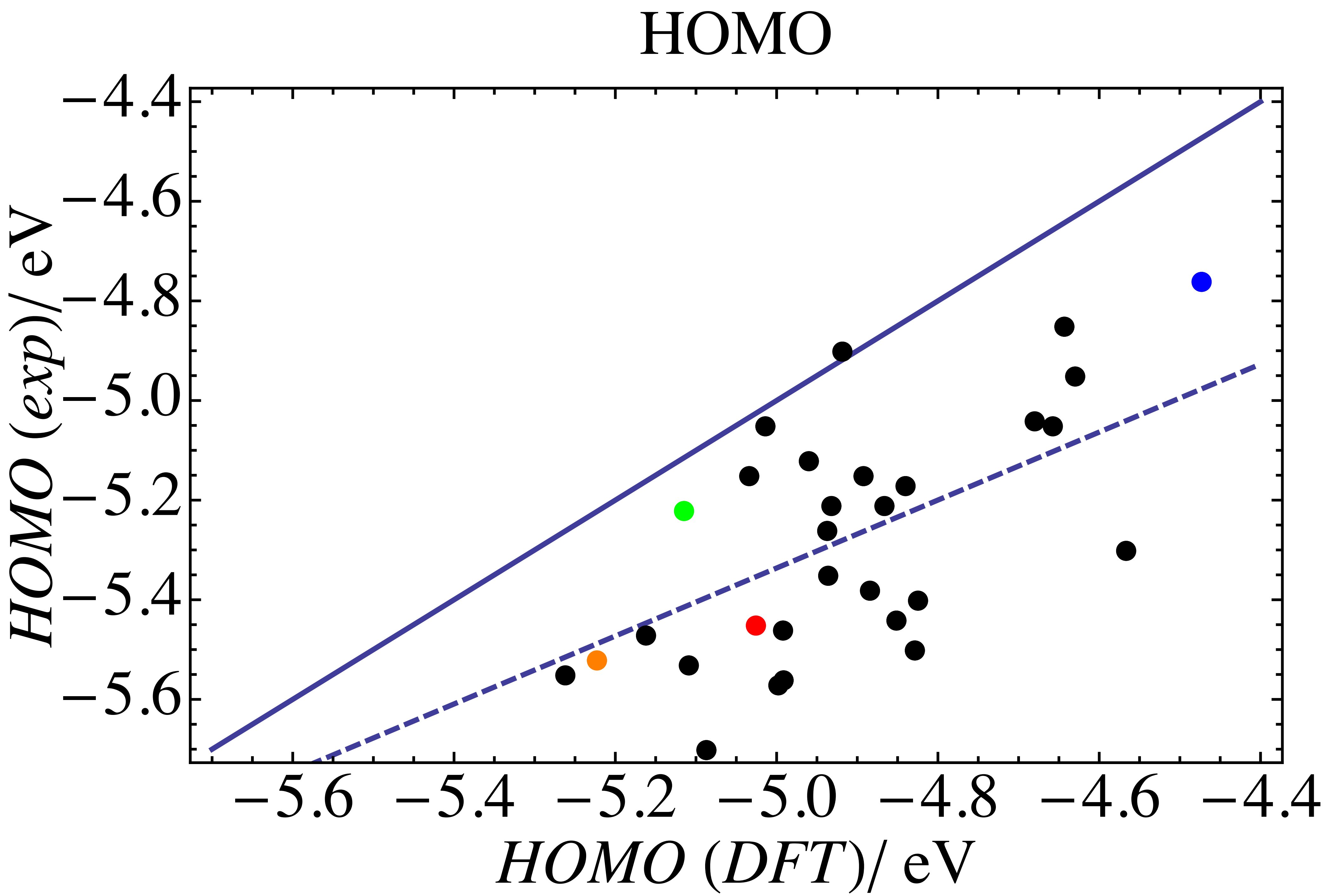
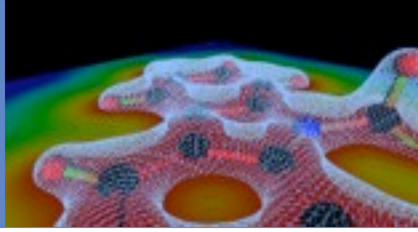




- Allows to select the top 10-20% candidates polymers



The G_0W_0 : performance

G_0W_0 :

$$\left(T + V_{ext} + V_H + V_{xc}\right)\left|\phi_n\right\rangle = \varepsilon_n \left|\phi_n\right\rangle$$

$$G = \sum_{n=1}^{\infty} \frac{\left|\phi_n\right\rangle\left\langle\phi_n\right|}{\omega - \varepsilon_n}$$

$$P = -iGG$$

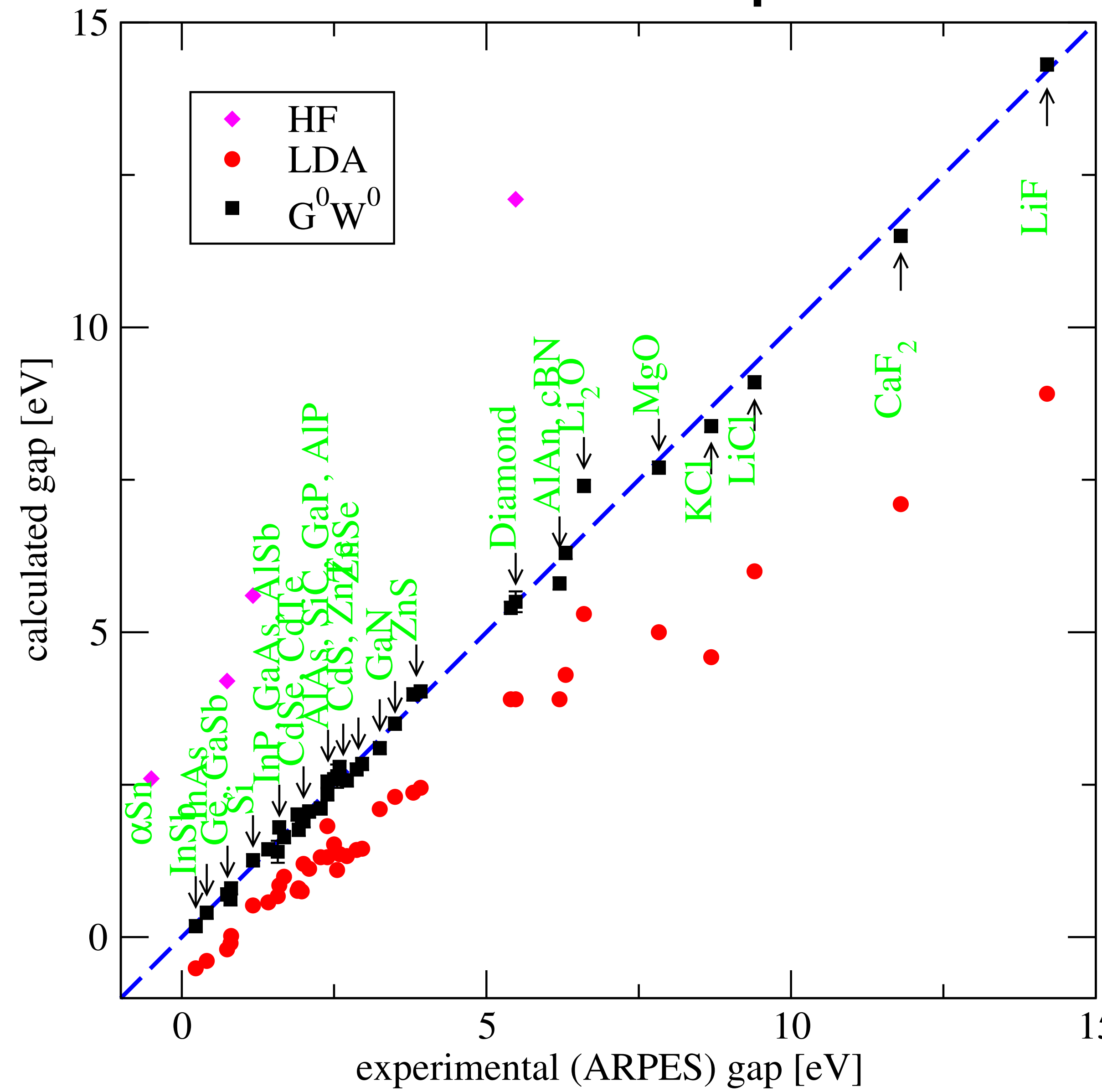
$$\epsilon = 1 - vP$$

$$W = \epsilon^{-1} v$$

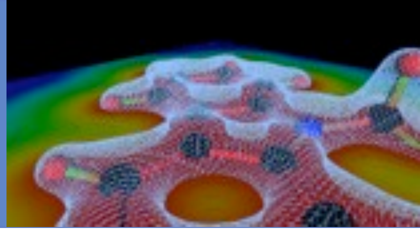
$$\Sigma = iGW$$

$$\epsilon_n \approx \varepsilon_n + \left\langle\phi_n\right|\Sigma - V_{xc}\left|\phi_n\right\rangle$$

LDA and G_0W_0 : precision



S. Faleev, PRL, 2004



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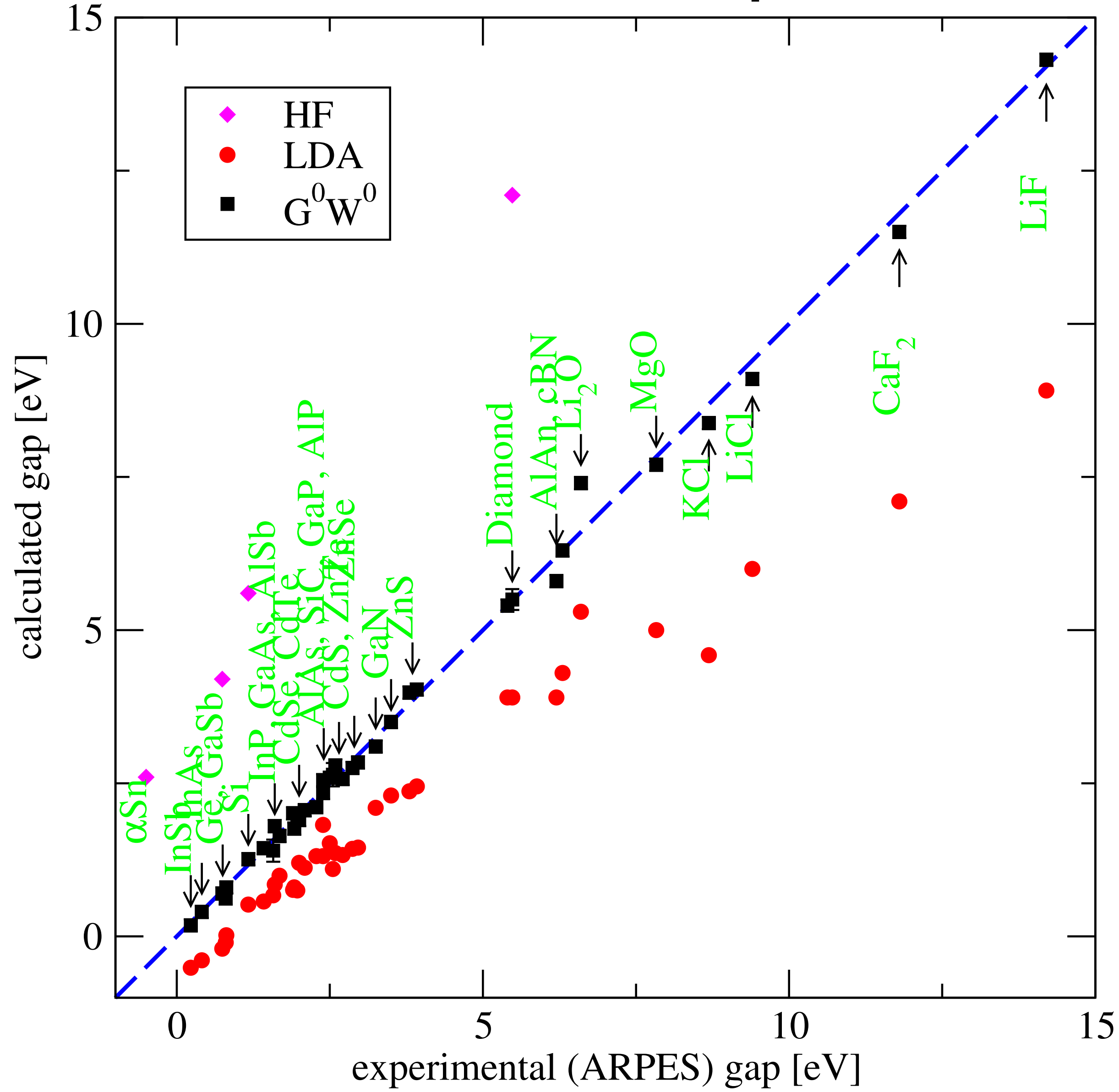

~ 10 atoms !

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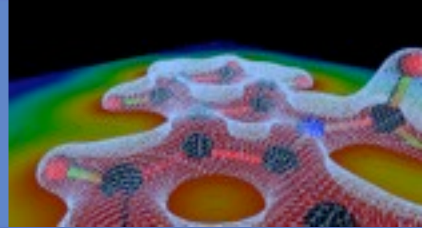

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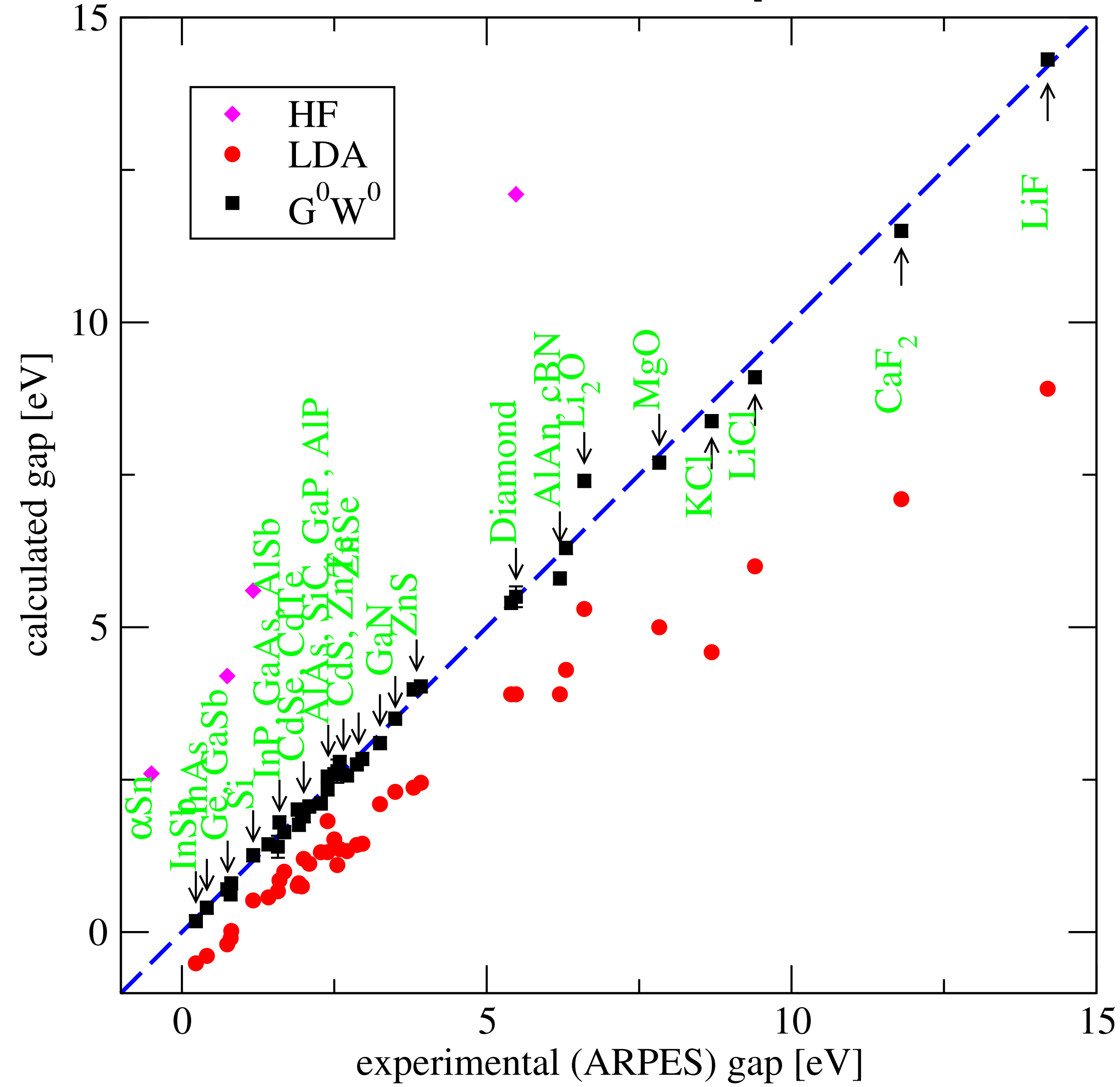

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The G_0W_0 : bottlenecks

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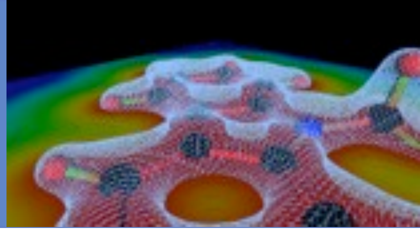

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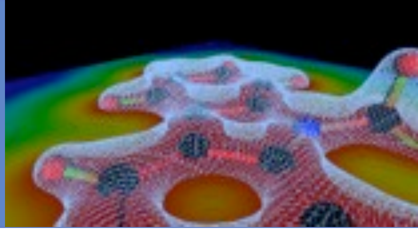

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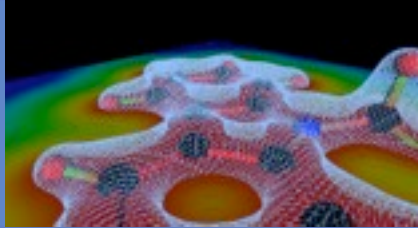

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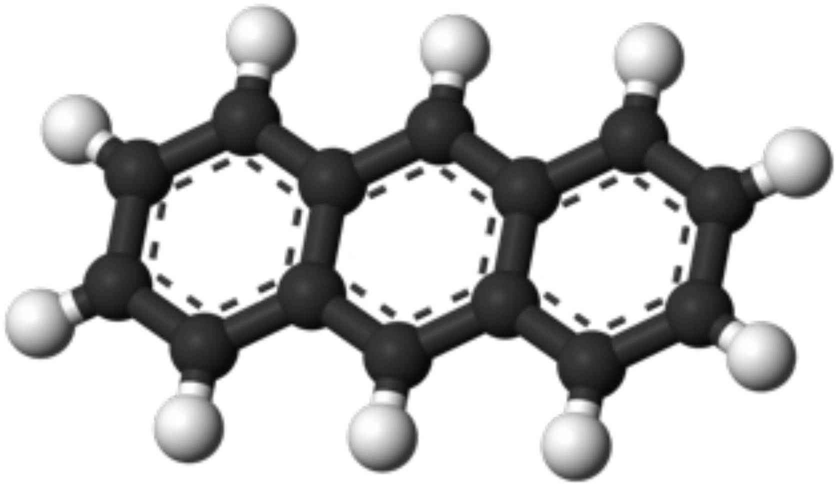
- inversion of $\epsilon \Rightarrow N^3$ operation (N = basis size)

The case of antracene



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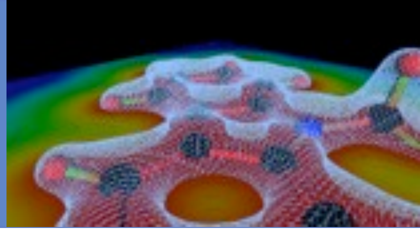

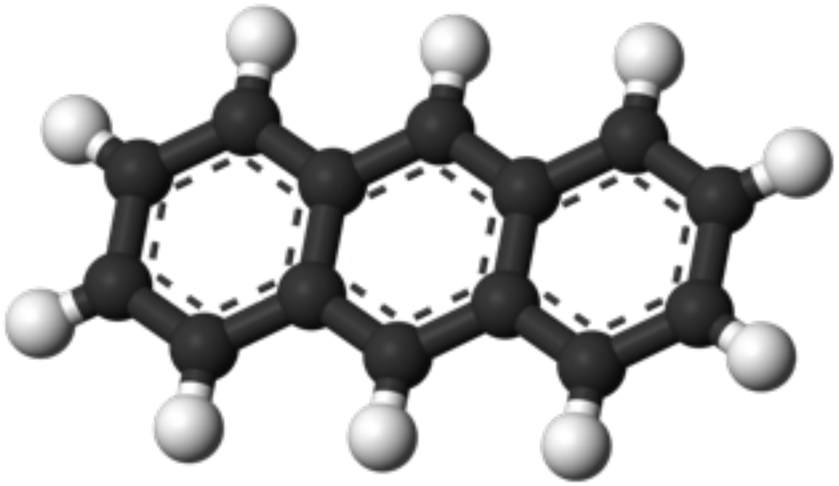

$N_v = 33$

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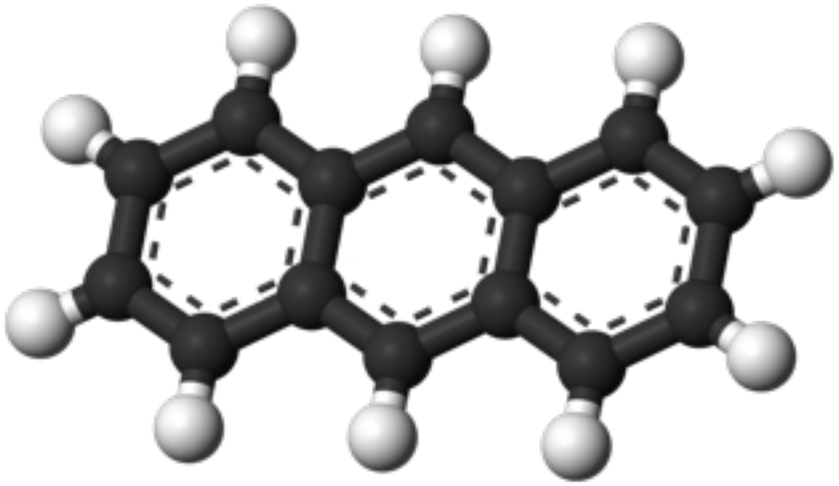

$N_v = 33$

- $N_c \sim 3000$ and $N_{basis} \sim 200\ 000$

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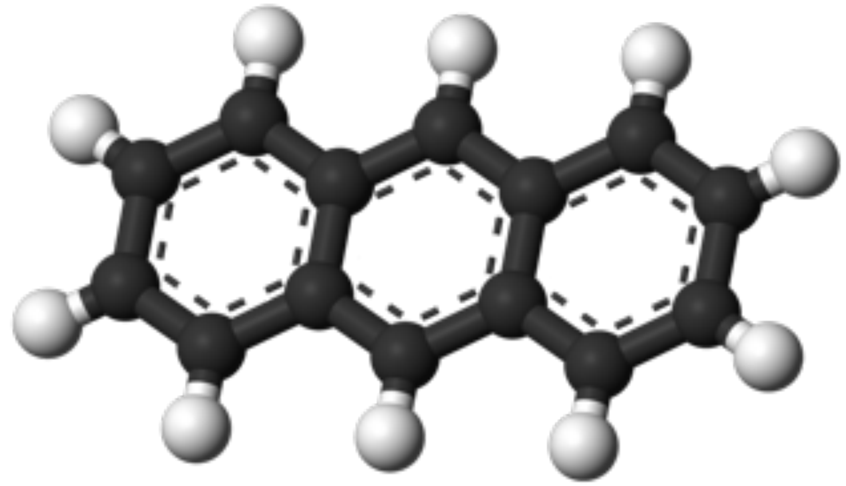

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➡ 10 Gb of RAM usage to store $\{|c\rangle\}$

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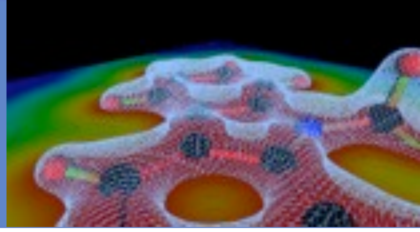

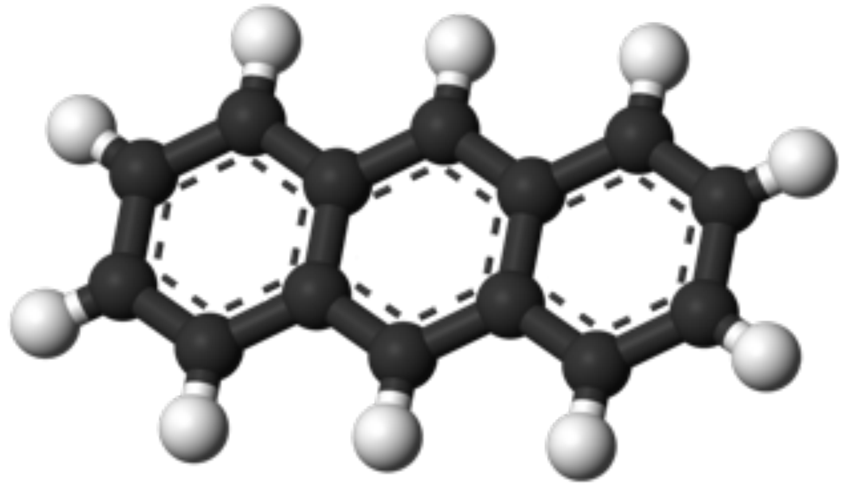

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- N_c ~ 3000 and N_{basis} ~ 200 000

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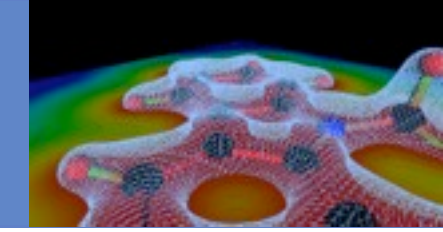

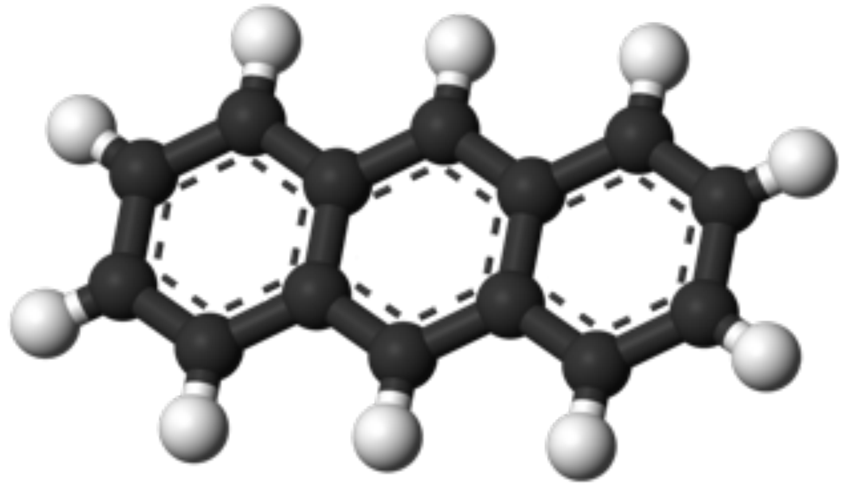

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- ϵ matrix ~ 7000 x 7000 planewaves

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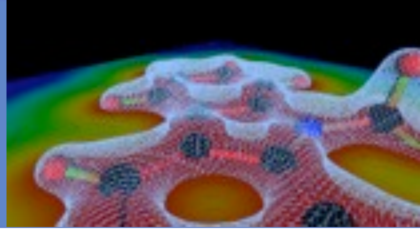

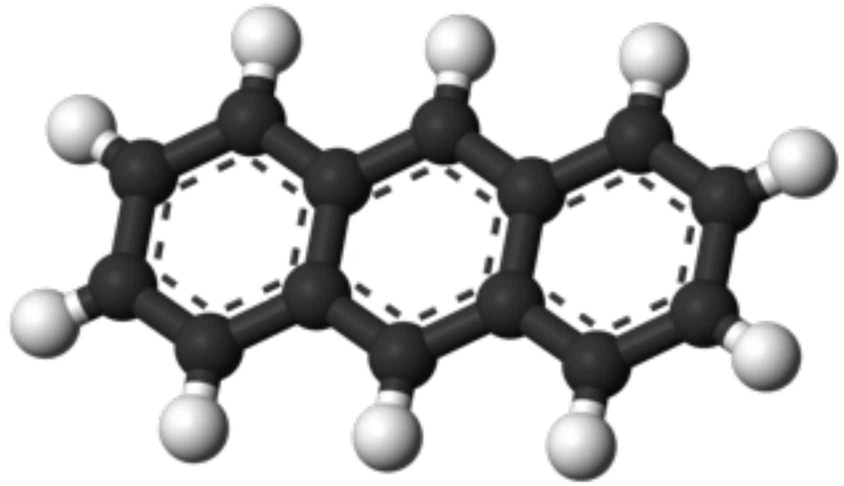

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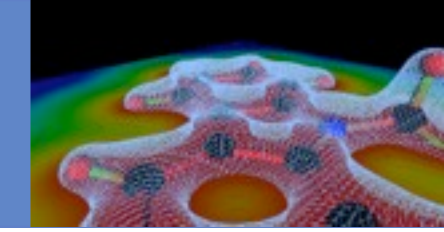

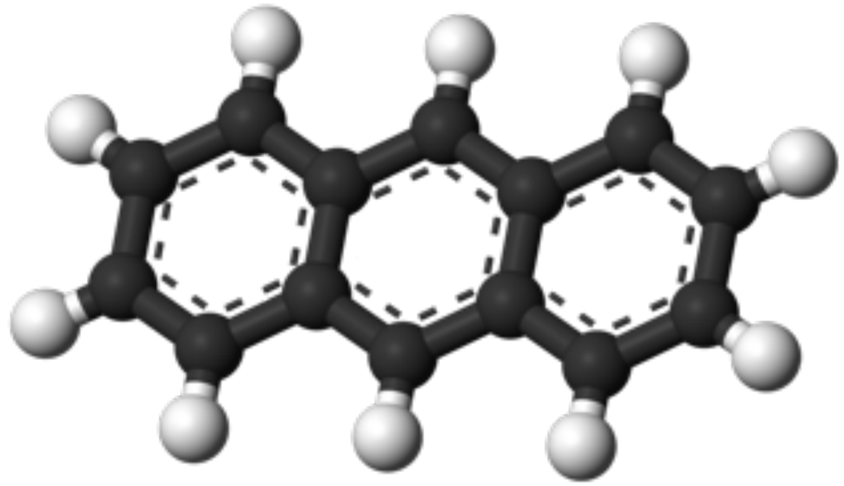

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➡ 1 Gb of RAM usage to store ϵ

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The plan

$$P = -iGG$$

$$\epsilon = 1 - vP$$

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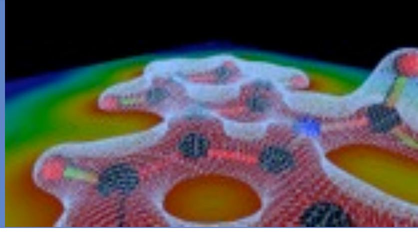
$$\Sigma = iGW$$

N_c : number of conduction states

$$G = \sum_{n=1}^{\infty \sim N_c} \frac{|n\rangle\langle n|}{\omega - \varepsilon_n}$$



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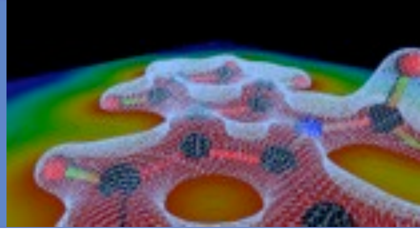

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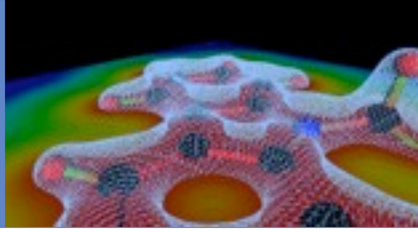
- summations → Sternheimer's equations

- planewaves basis → Lanczos basis

Sternheimer's equation

- $$P\left|\psi\right\rangle=\sum_v\left|v\right\rangle\left(\sum_c\left|c\right\rangle\frac{1}{\omega-(\varepsilon_c-\varepsilon_v)}-\frac{1}{\omega+(\varepsilon_c-\varepsilon_v)}\left\langle c\right|\left\langle v\right|\right)\left|\psi\right\rangle$$

1) $P = -iGG$ Sternheimer's equation



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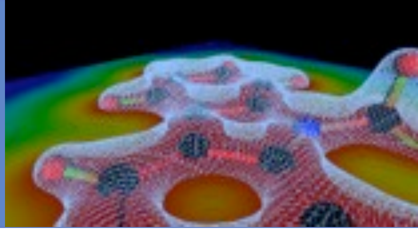

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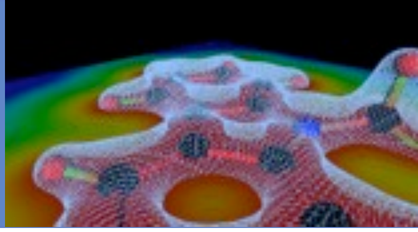

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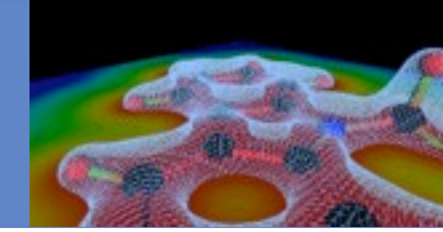

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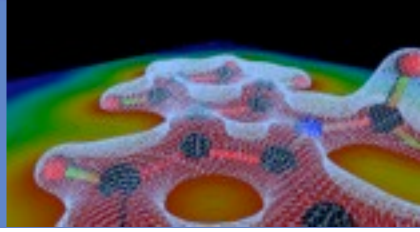

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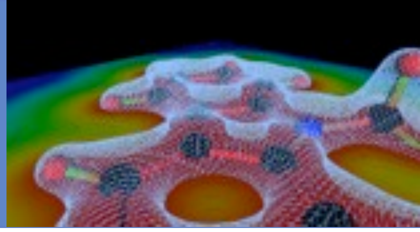

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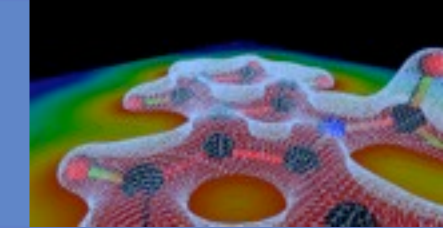

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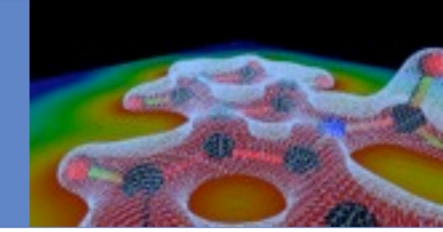
$$|\phi_v^-\rangle \equiv \sum_c \frac{|c\rangle\langle c|}{\omega-(\varepsilon_c-\varepsilon_v)} |v\rangle|\psi\rangle = \frac{1}{\omega - H + \varepsilon_v}\mathcal{P}_c |v\rangle|\psi\rangle$$

1) $P = -iGG$ Sternheimer's equation



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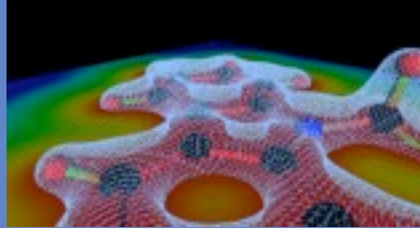
Sternheimer's equation



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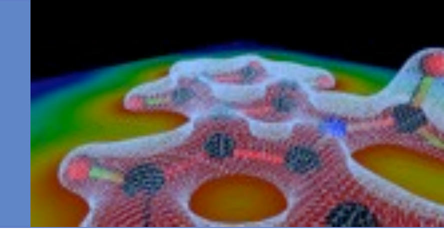

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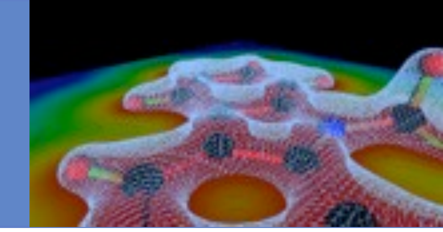

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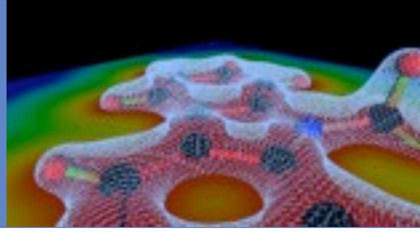

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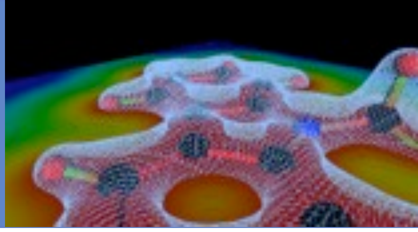

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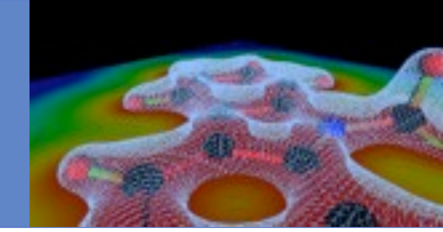

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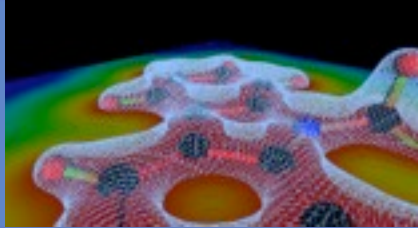

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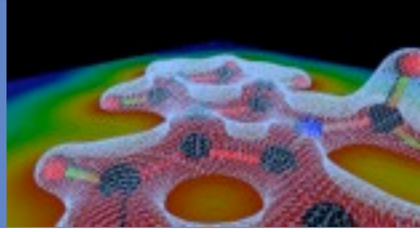

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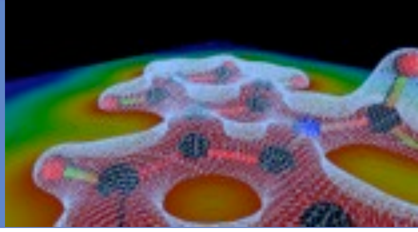

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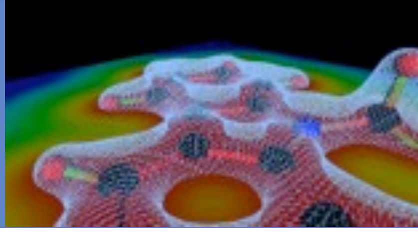

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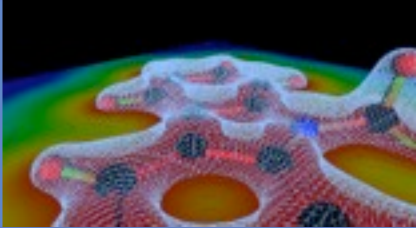

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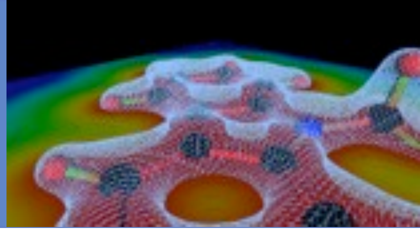

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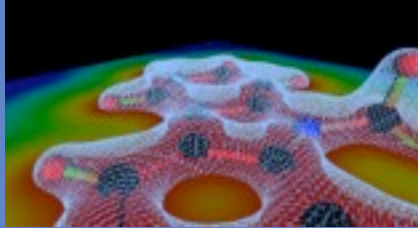

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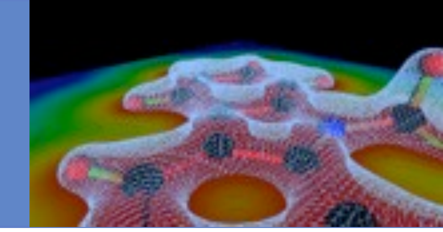

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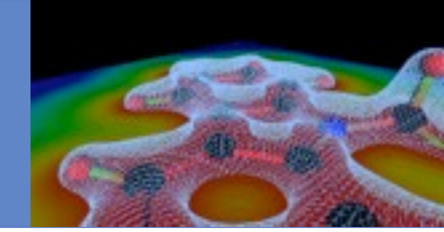

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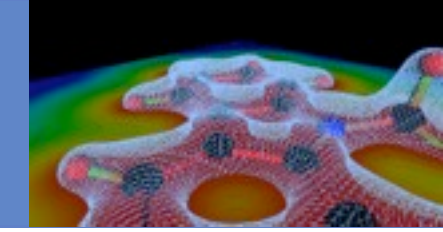

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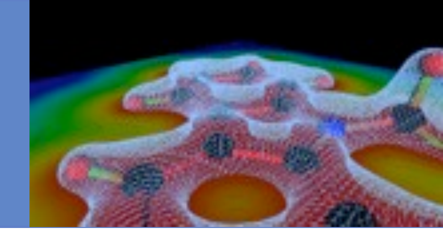

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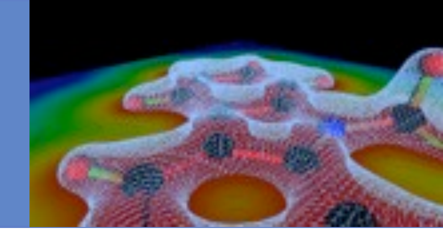

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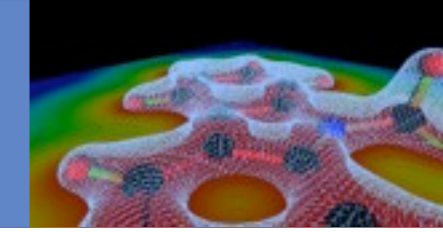
Sternheimer's equation



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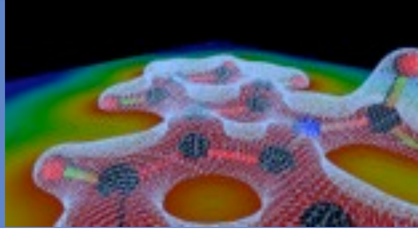

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Lanczos algorithm

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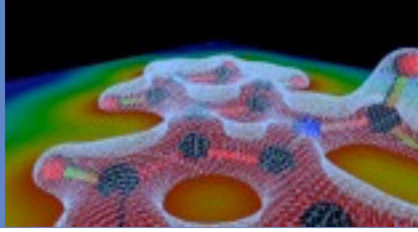
- Need a basis: $\{|q\rangle\}$

Lanczos algorithm



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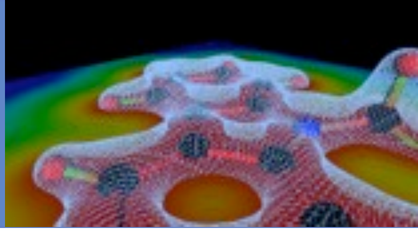

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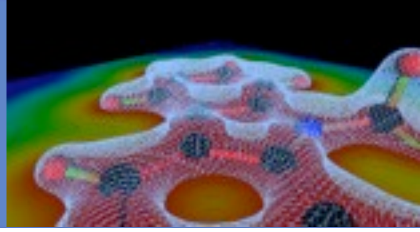

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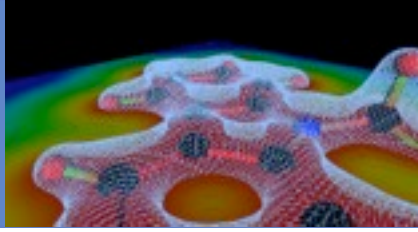
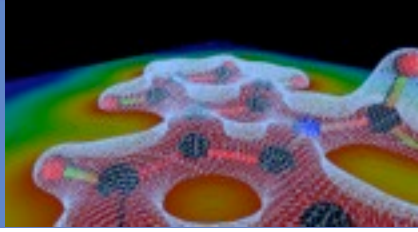
  - Is small, e.g.
    $\Rightarrow$ NOT planewaves
  - Is easy to compute
    $\Rightarrow$ NOT $\{|n\rangle\}$

Lanczos algorithm



Lanczos algorithm



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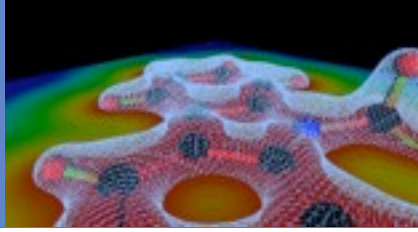
Lanczos algorithm



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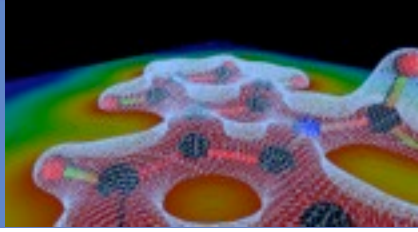
Lanczos algorithm



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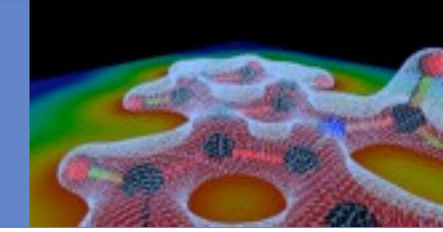
    $\rightarrow$ biggest eigenvalues pop out

Lanczos algorithm



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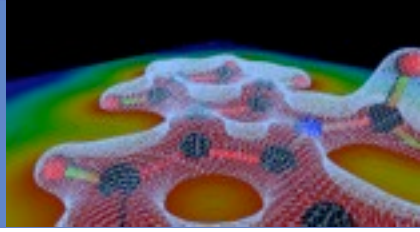

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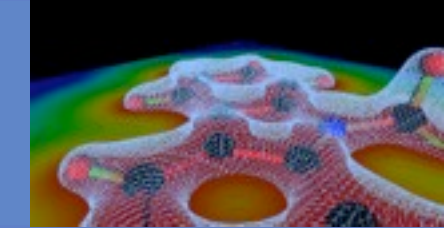
2) $\Sigma = iGW$ Lanczos algorithm



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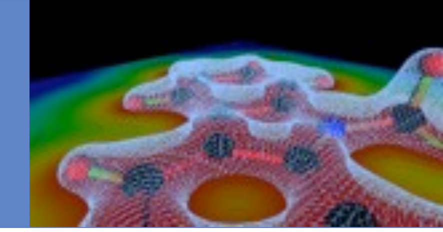
Lanczos algorithm



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  - Orthonormalize : $\{|q\rangle\}$
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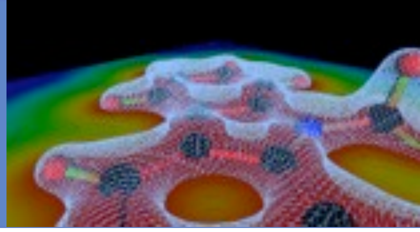
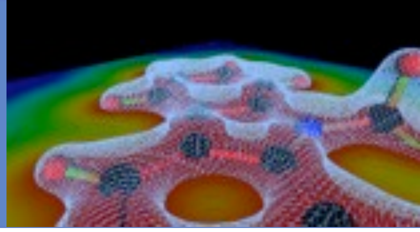
  - Don’t pay all orthogonalization
  - Obtain ϵ for free

3) $W = \epsilon^{-1} v$

Lanczos algorithm

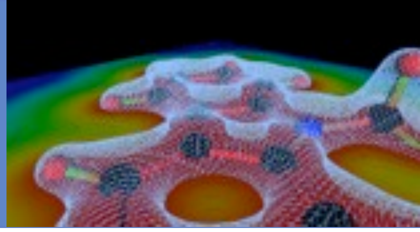


3) $W = \epsilon^{-1} v$ — Lanczos algorithm



$$\langle m | \Sigma^c | m \rangle = \frac{i}{2\pi} \int_{-\infty}^{+\infty} d\omega \sum_q \langle q | \left[\epsilon^{-1} - 1 \right] (\ldots) | q \rangle$$

3) $W = \epsilon^{-1} v$ Lanczos algorithm



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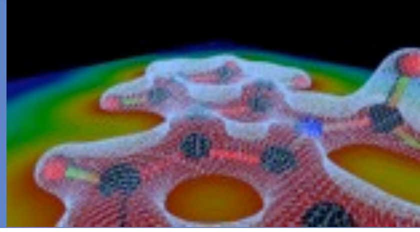

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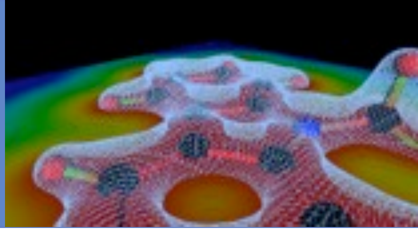

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- We have a small $\{|q\rangle\}$

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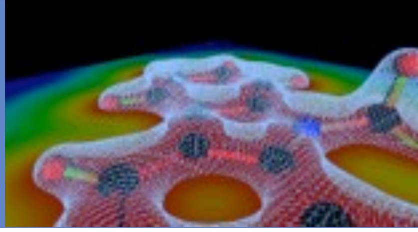

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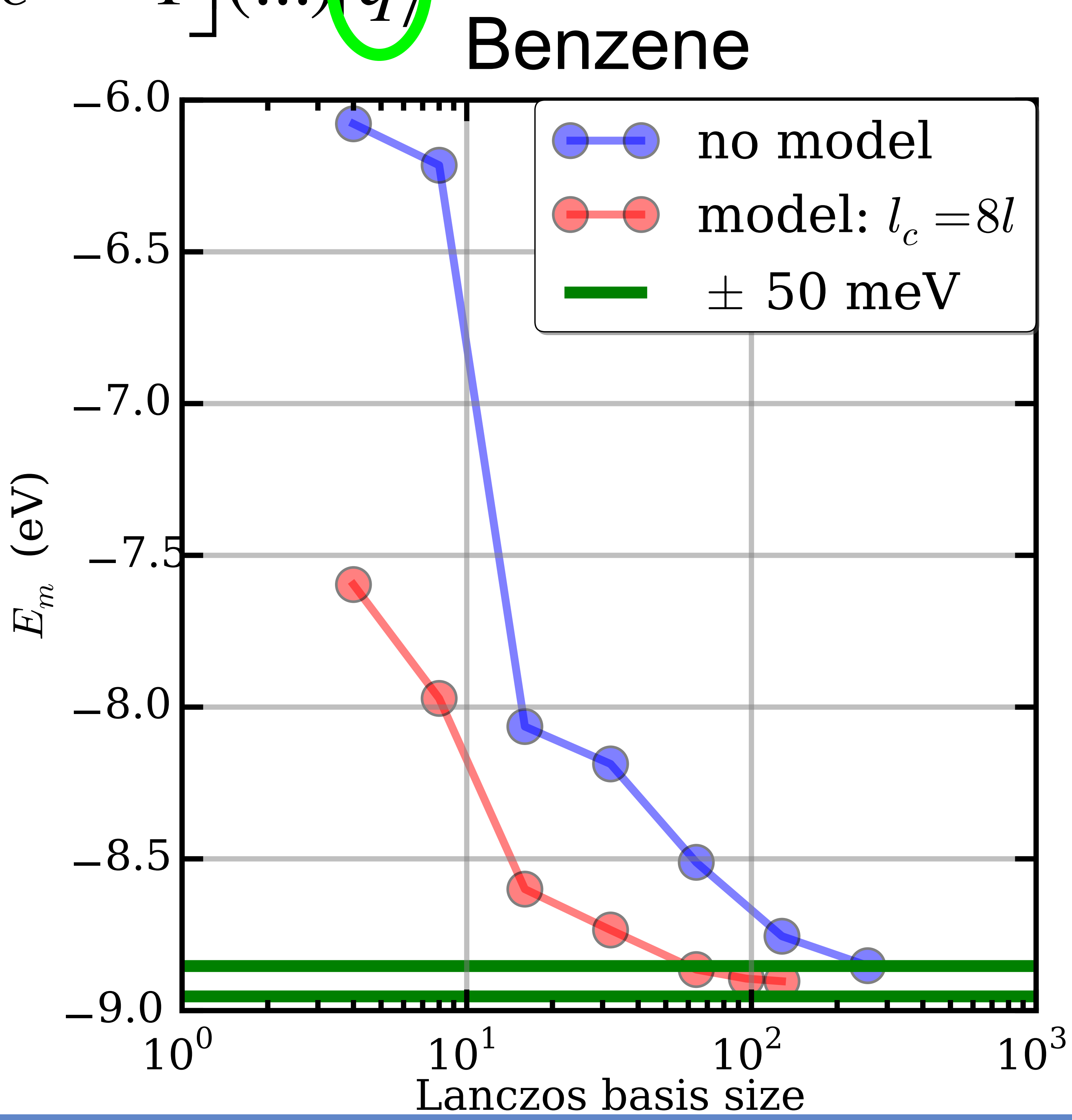

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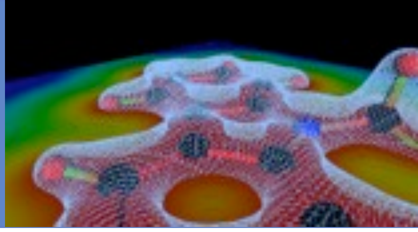

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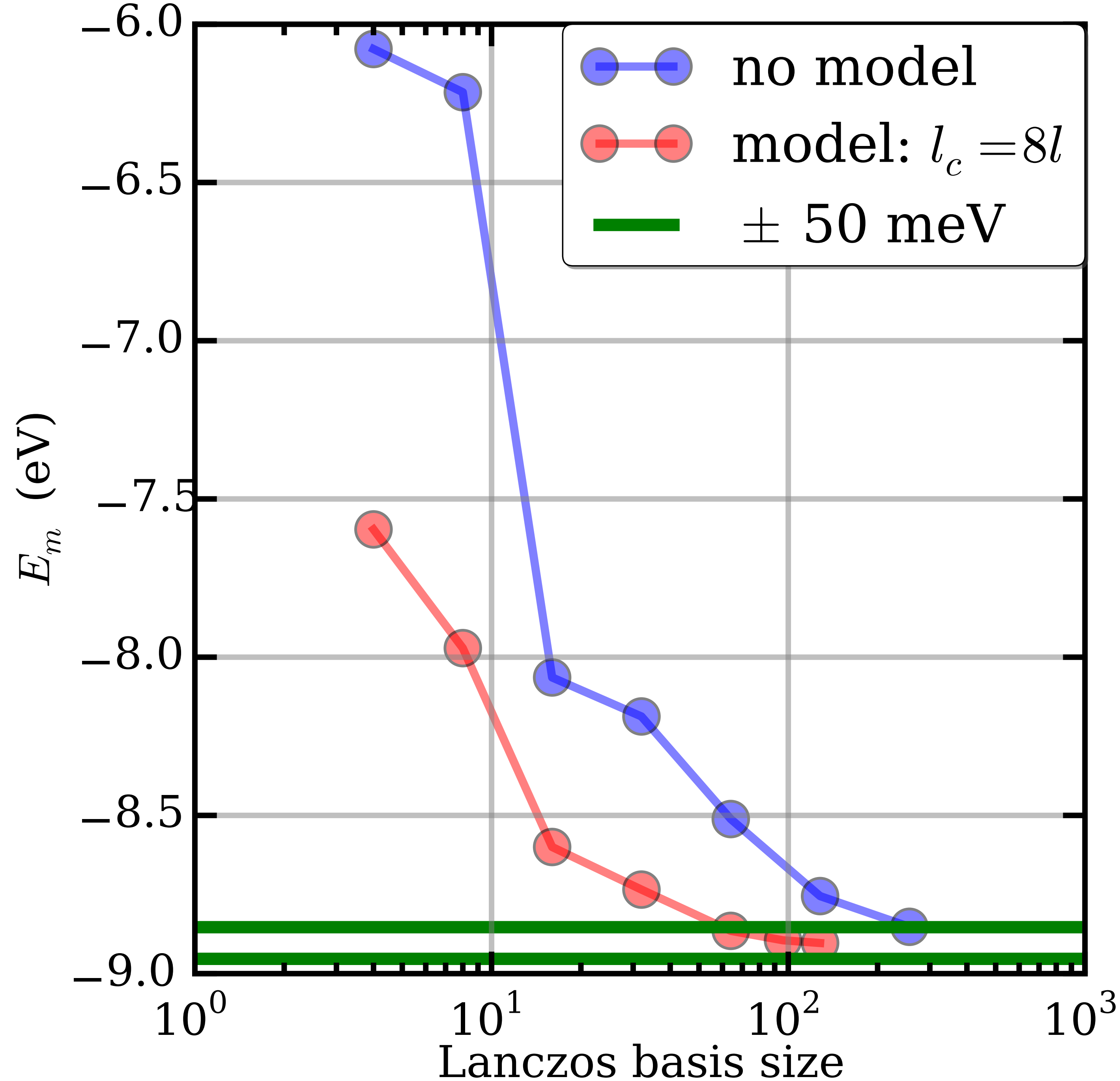
- We have a small $\{|q\rangle\}$

  $\rightarrow \epsilon^{-1}$ ~ free

Benzene



Performance

Performance

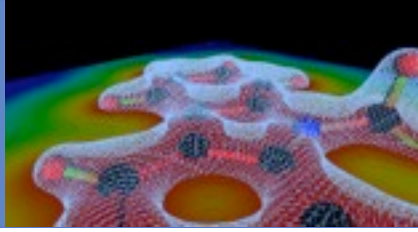
$$\cancel{\sum_n}$$

Performance

$$\cancel{\sum_{n}}$$

$$\cancel{\epsilon^{-1}}$$

(smaller)



Performance

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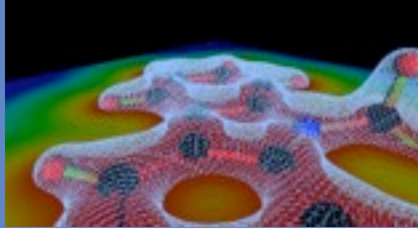
$$\cancel{\epsilon^{-1}}$$

(smaller)

$$\boxed{A|x\rangle = |b\rangle}$$



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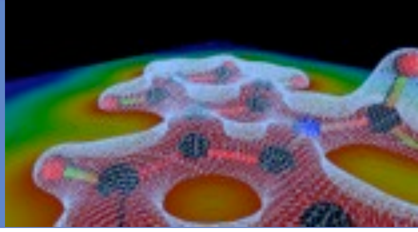
$$\boxed{\{|q\rangle\}}$$



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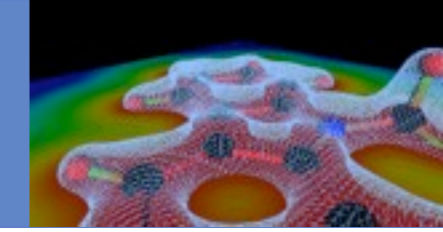
Faster ?

Performance



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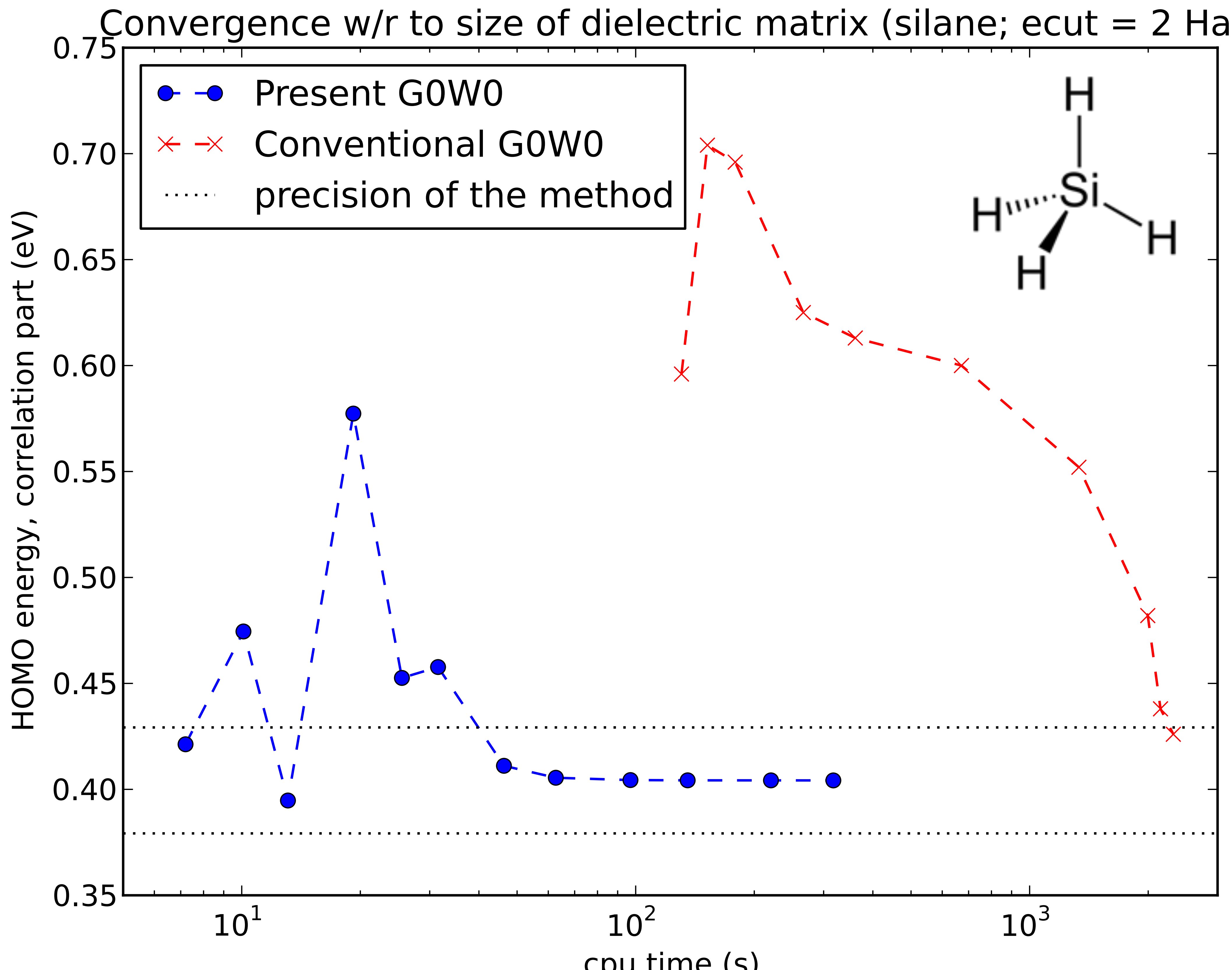

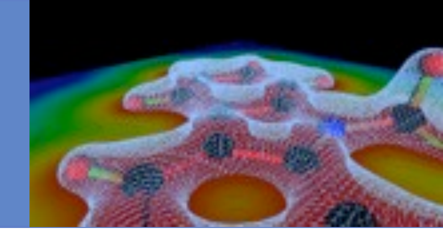

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$\cancel{\epsilon^{-1}}$ (smaller)

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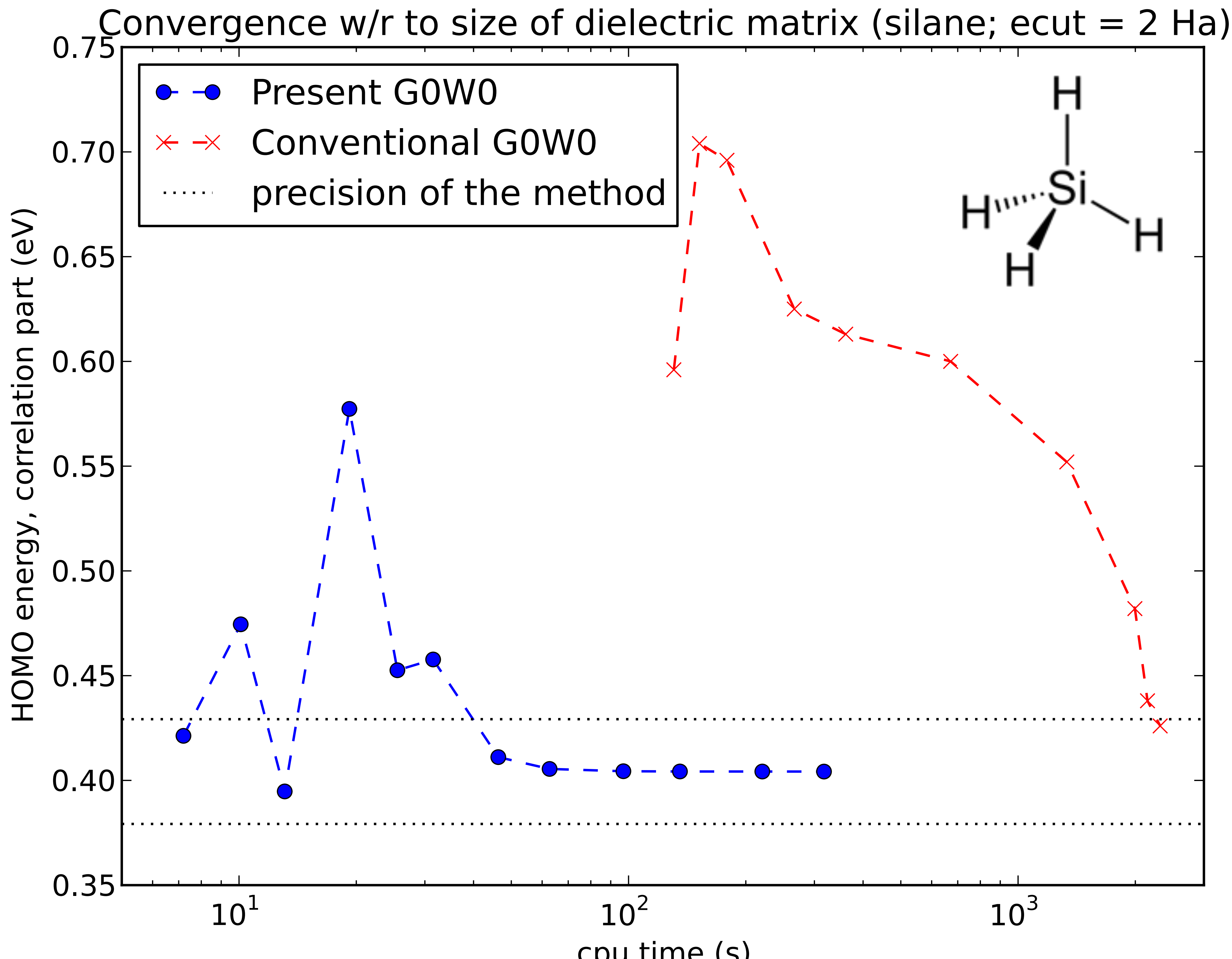
Faster ?

Yes!



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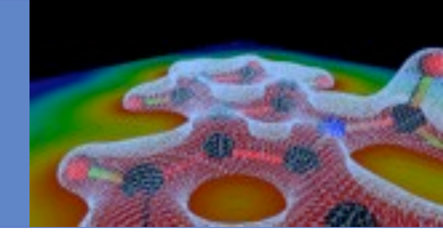

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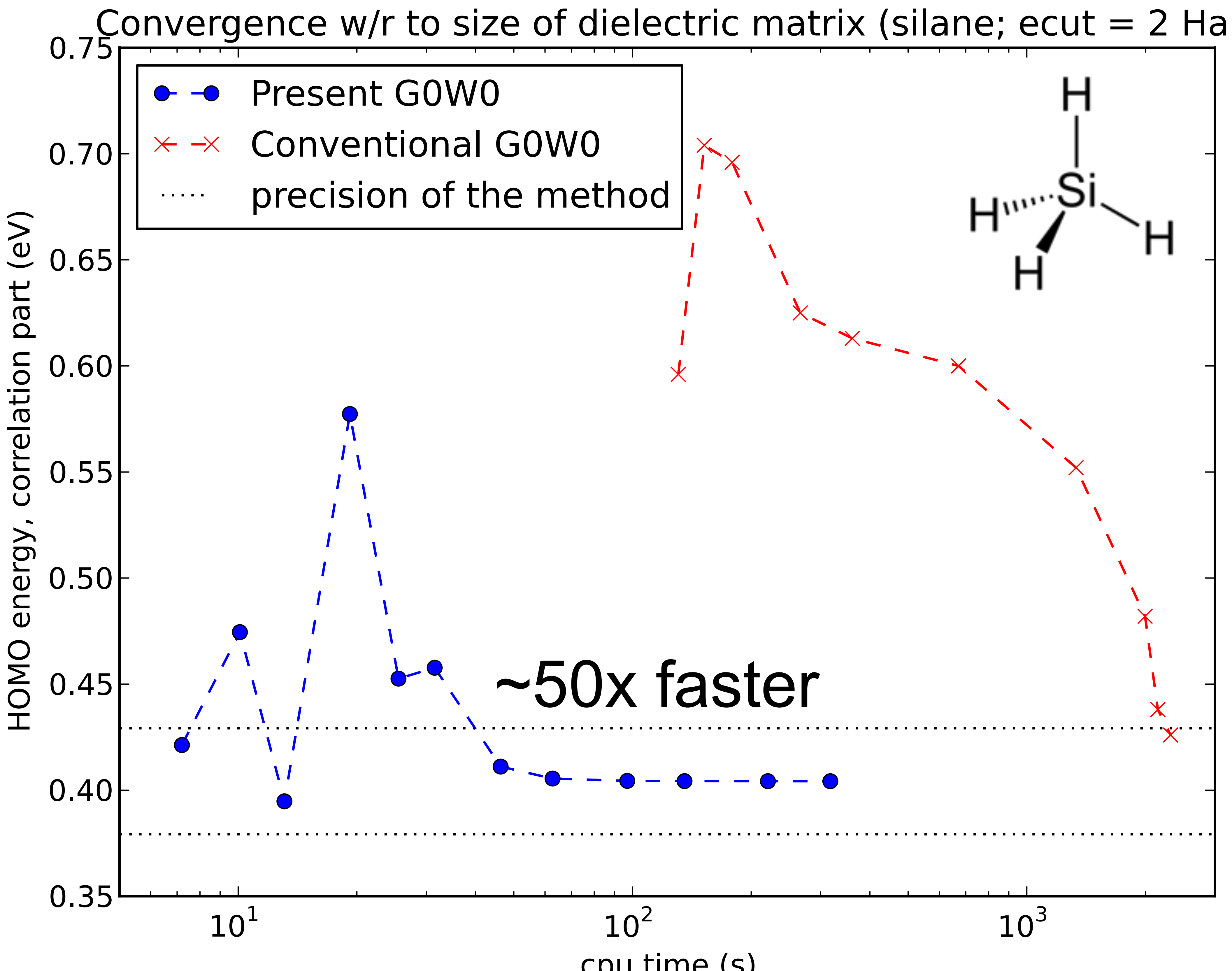

Preliminary results

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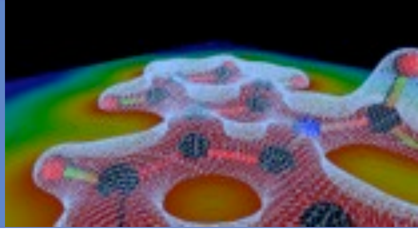
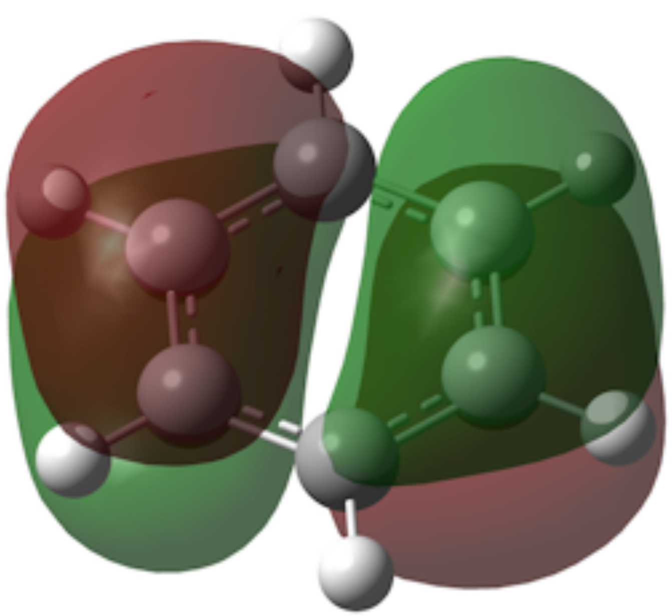


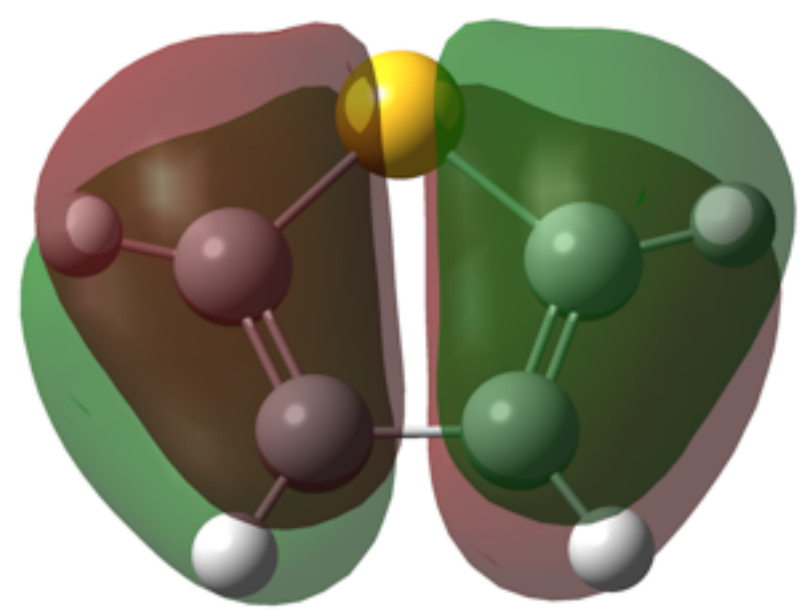
- Working in reals systems?

Preliminary results

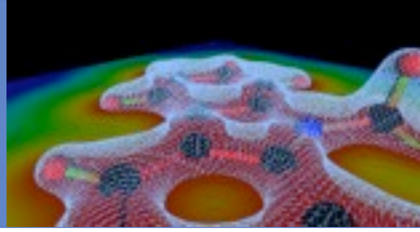
- Working in reals systems?

	HOMO (eV)		
	LDA	**G_0W_0**	**Exp.**
Benzene	-6.51	-9.22	-9.30
Thiophene	-6.05	-8.94	-8.85





Conclusion



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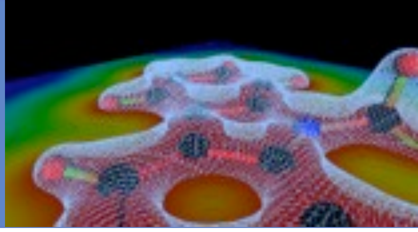

- Bottleneck assessed :

Conclusion

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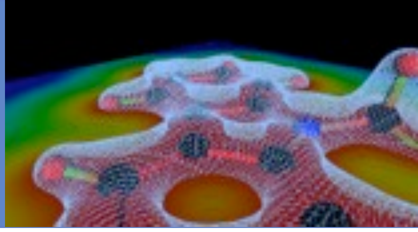

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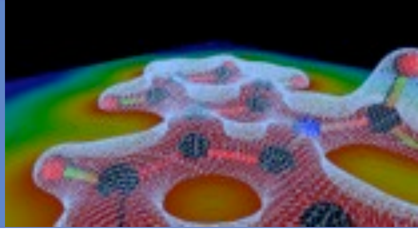

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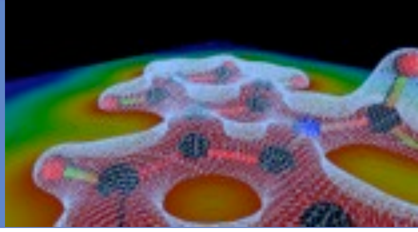
- Future work :

Conclusion



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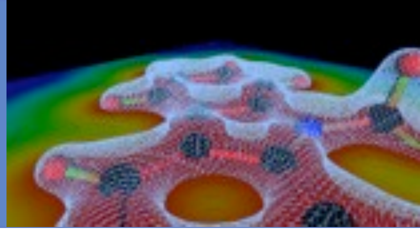

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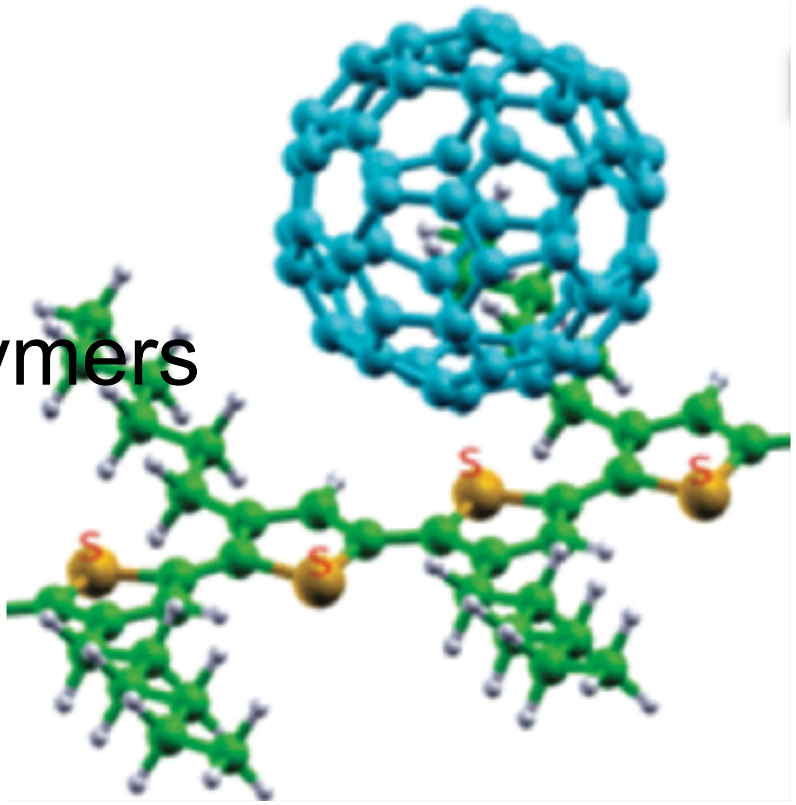

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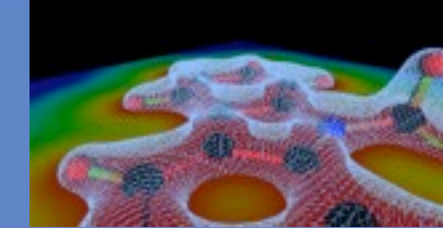

Thank you!

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Acknowledgments

Michel Côté's group

Thank you!

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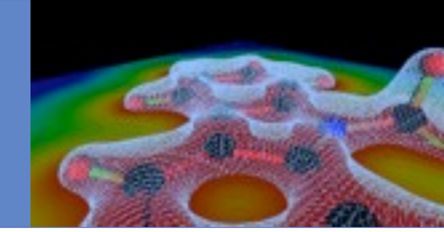

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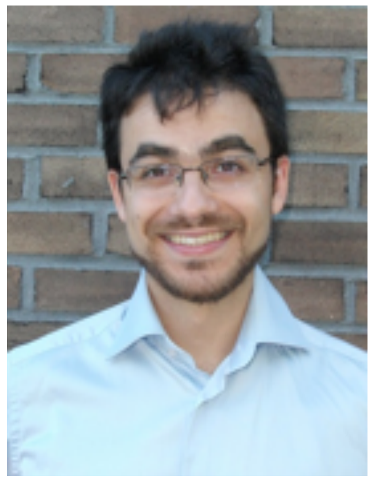

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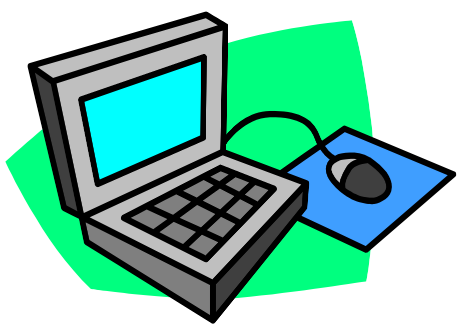

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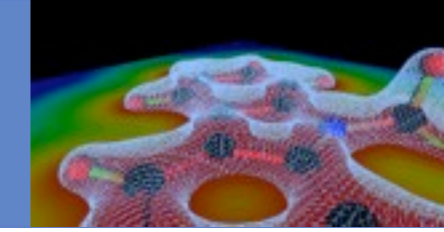

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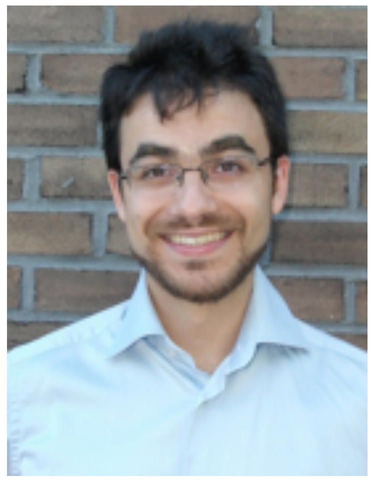

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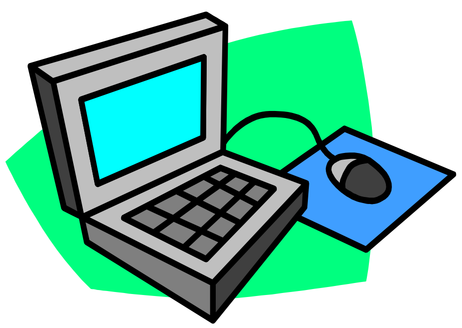

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