Hot electron dynamics in graphene –a linear-scaling atomistic approach

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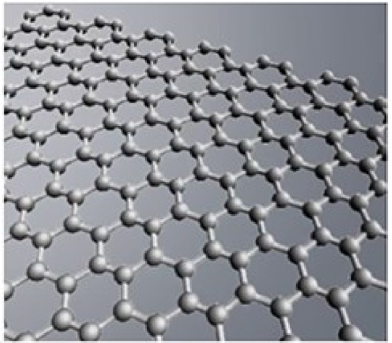
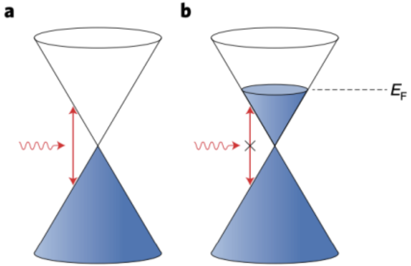
Graphene, novel devices and sensing technologies

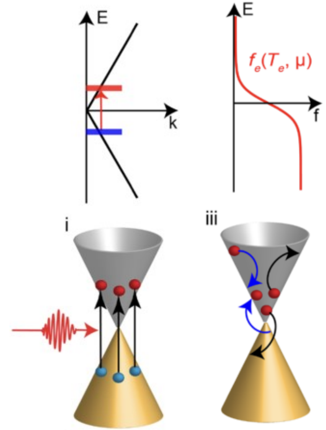


Carrier density control



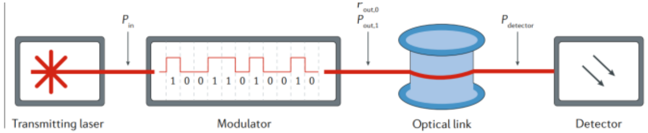
Ferrari, A. C. Nature Photonics, 15(7), 488-490 (2021).

Efficient carrier heating



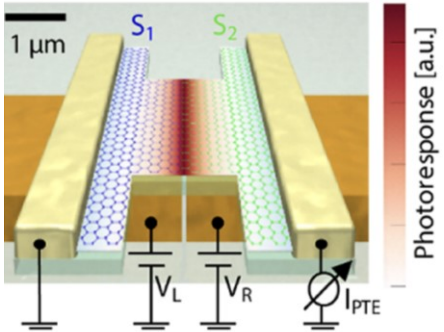
Chen, Y., et al. Science advances, 5(11), eaax9958 (2019).

Optical modulators



Romagnoli, M. , et al., Nature Reviews Materials, 3(10), 392-414 (2018)

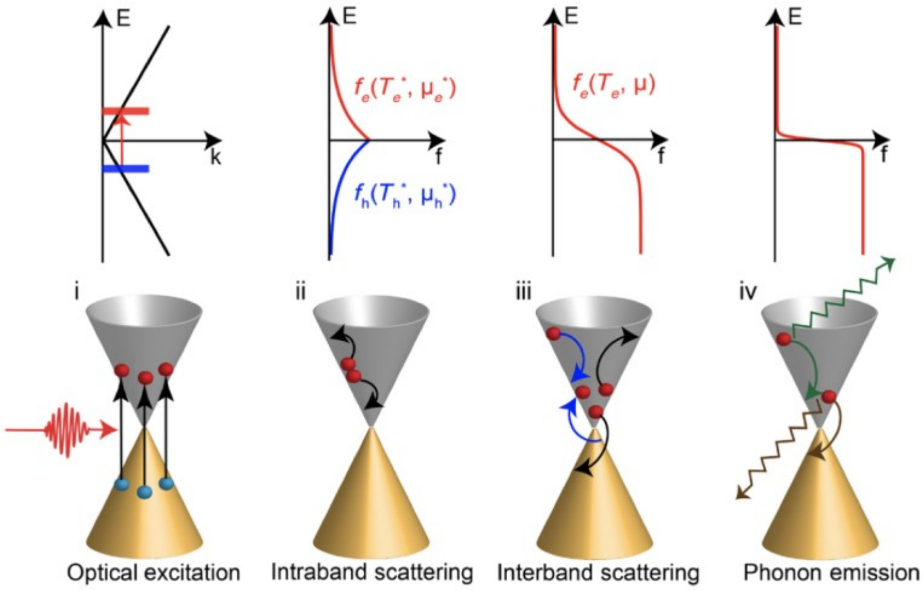
Thz sensors



Castilla, S.,et al. Nano letters, 19(5), 2765-2773 (2019).

Hot Carrier Relaxation in Graphene

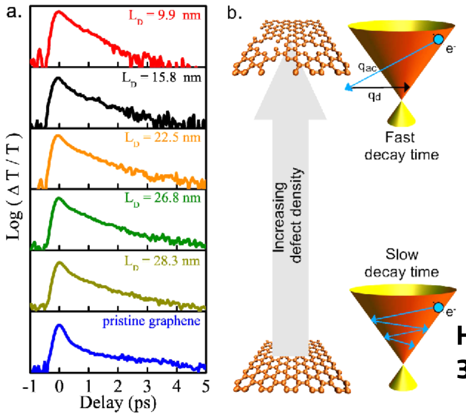
$\sim 10 - 100$ fs $\quad\quad \sim 1 - 100$ ps

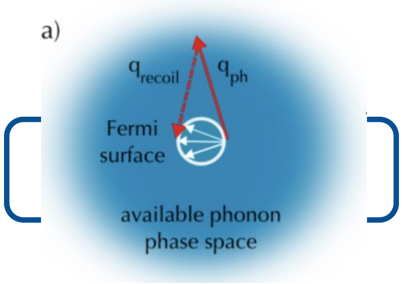


H_{el-el} $\quad\quad$ H_{el-ph}

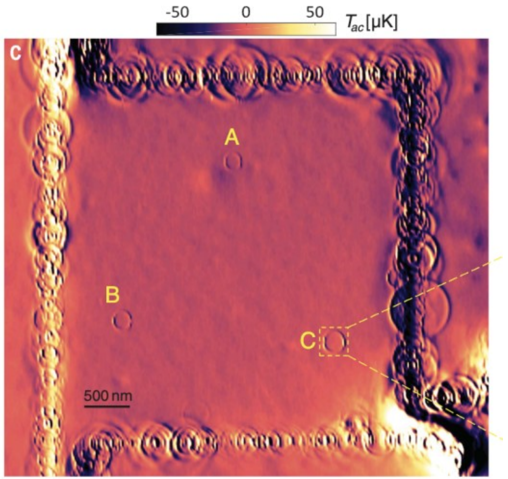
Song, J. C., Reizer, M. Y., & Levitov, L. S. Physical review letters, 109(10), 106602 (2012).

Alencar, T. V., et al. Nano letters, 14(10), 5621-5624 (2014).





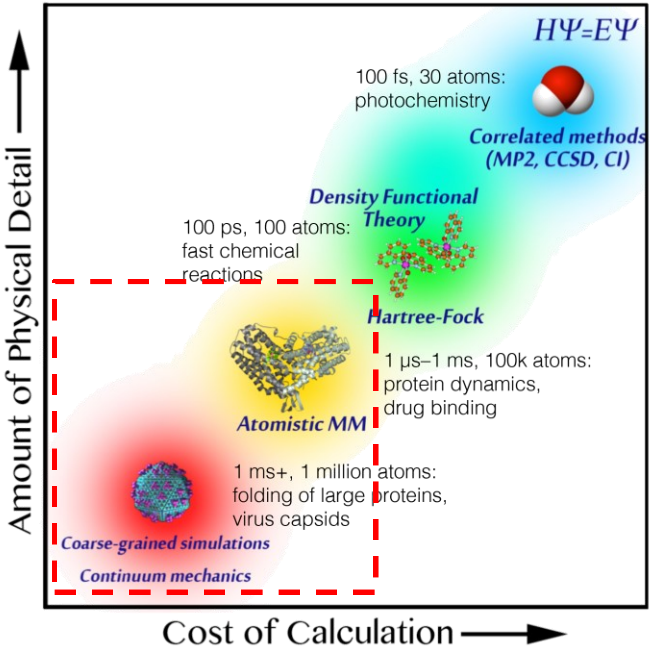
Halbertal, D., et al. Science, 358(6368), 1303-1306 (2017).

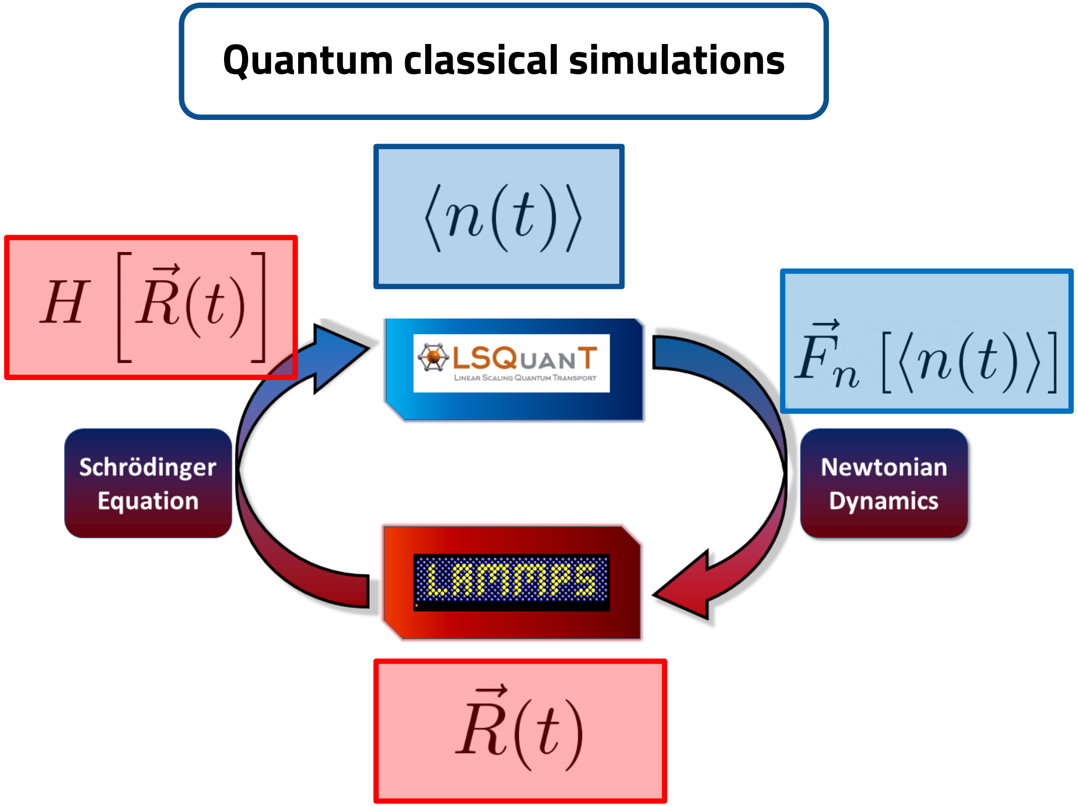


Resonant cooling

We need tools for dealing with disordered systems in these regimes!

Quantum-classical simulations





Linear scaling quantum transport methodologies

- **Chebyshev polynomial expansion of spectral functions**

$$f(\alpha, \beta, H) = g_n \mu_n T_n(H)$$

$$\mu_n = \frac{2}{\pi} \int_{-1}^{1} \frac{f(\tilde{\varepsilon}) T_n(\tilde{\varepsilon})}{\sqrt{(1-\tilde{\varepsilon})}} d\tilde{\varepsilon}$$

- **Spectral functions of interest**

$$U(t) = exp\left(\frac{-i}{\hbar} \int dt' H(t')\right) \approx \prod_{n=0}^{N} exp\left(\frac{-i}{\hbar} H_n \Delta t\right)$$

$$f(\varepsilon, \mu, T) = \frac{1}{e^{(\varepsilon-\mu)/K_B T} + 1}$$

$$D(\varepsilon) = \delta(H - \varepsilon)$$

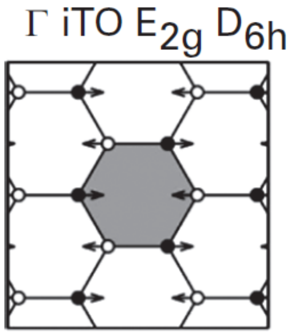
$$\langle n(t) \rangle = \frac{Tr\langle U(t)^\dagger f(H(0), \mu, T) U(t) \delta(H(t) - \varepsilon) \rangle}{Tr\langle \delta(H(t) - \varepsilon) \rangle}$$

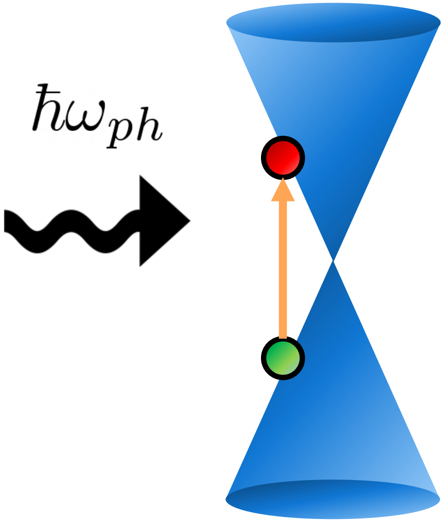
Fan, Z., et al. Physics Reports, 903, 1-69 (2021).

Ordejón, P. Computational materials science, 12(3), 157-191 (1998).

Non-perturbative phonon absorption in graphene

- Optical phonon absorption in graphene





$$H = \sum_{\langle i,j \rangle} t(\vec{r}_{ij}) b_j^\dagger a_i + h.c$$

$$t(\vec{r}_{ij}) = t_0 \exp(-3.37(l_{ij}/a_0 - 1))$$

$$\langle n(t) \rangle = \frac{Tr\langle U(t)^\dagger f(H(0), \mu, T) U(t) \delta(H(t) - \varepsilon) \rangle}{Tr\langle \delta(H(t) - \varepsilon) \rangle}$$

$$U(\Delta t) = exp\left(\frac{-i}{\hbar} H[R(t)] \Delta t\right)$$

$$A = \sqrt{\frac{\hbar^2}{2 M_C \hbar\omega_{ph}}}$$

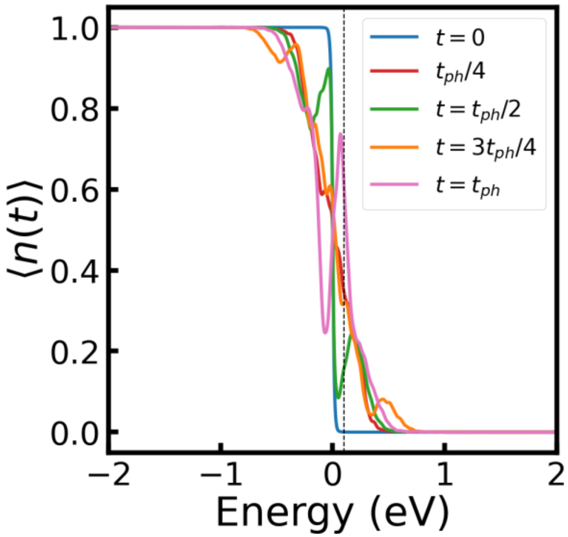
$$\hbar\omega_{ph} \approx 0.2 \text{ eV}$$

Avouris, P., Heinz, T. F., & Low, T. (Eds.). Cambridge University Press (2017).

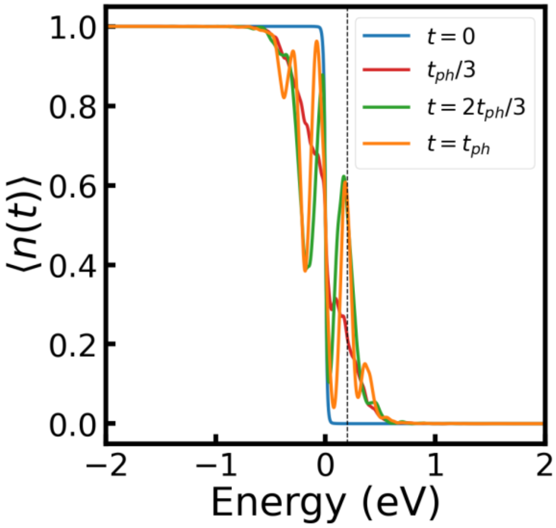
Non-perturbative phonon absorption in graphene

- **Optical phonon absorption in graphene**

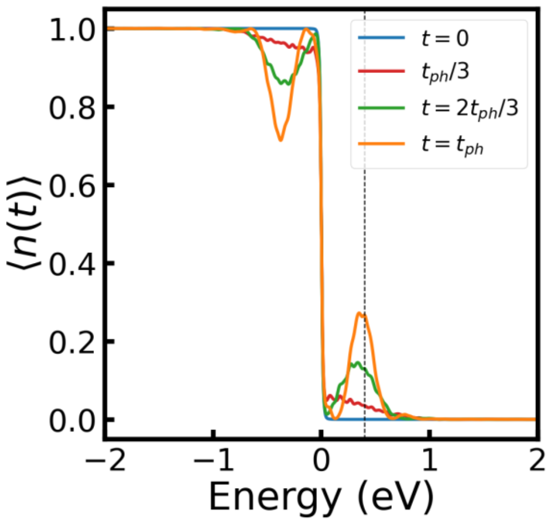
$\hbar\omega_{ph} = 0.2$ eV



$\hbar\omega_{ph} = 0.4$ eV



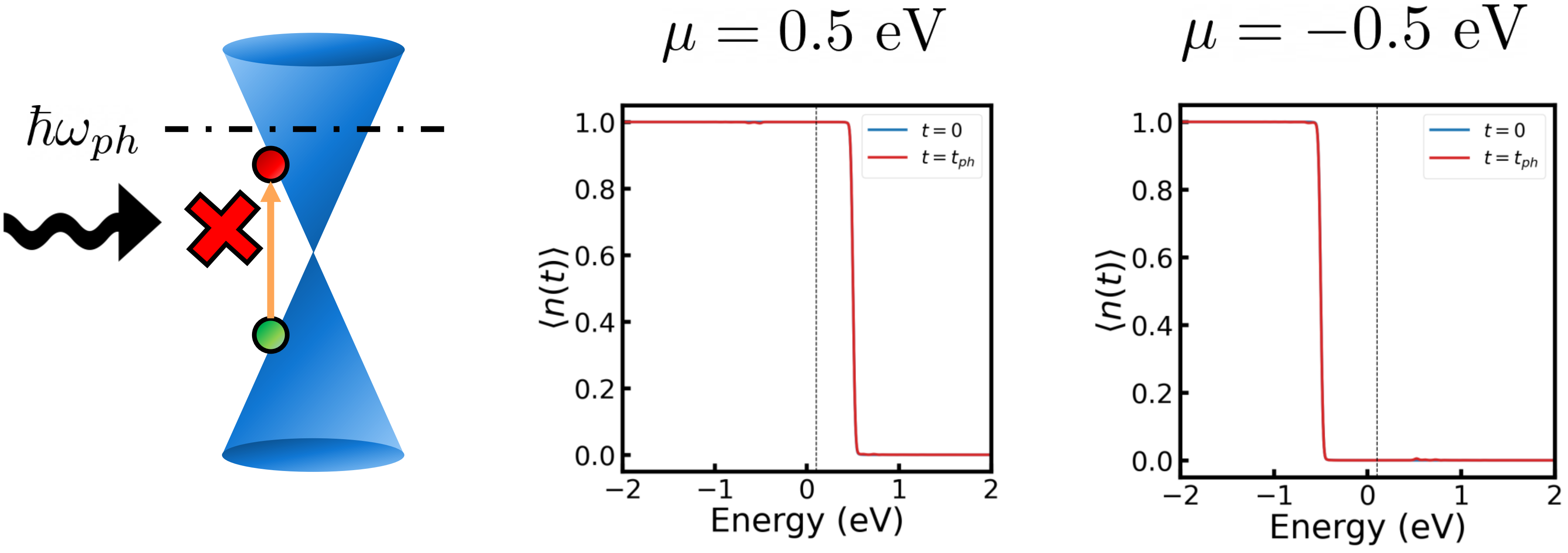
$\hbar\omega_{ph} = 0.8$ eV



$N \approx 4.19 \times 10^6$

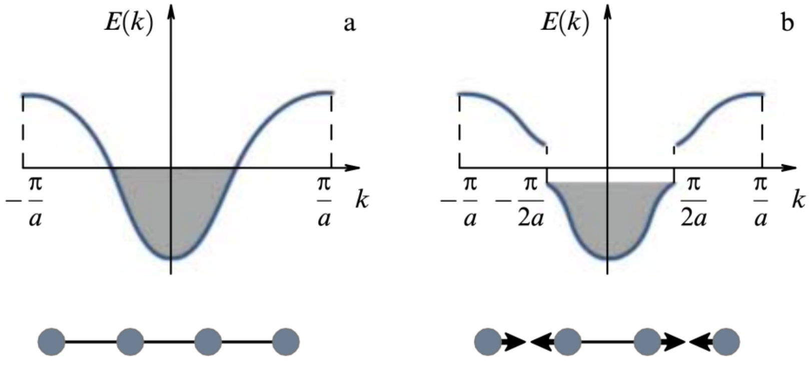
Non-perturbative phonon absorption in graphene

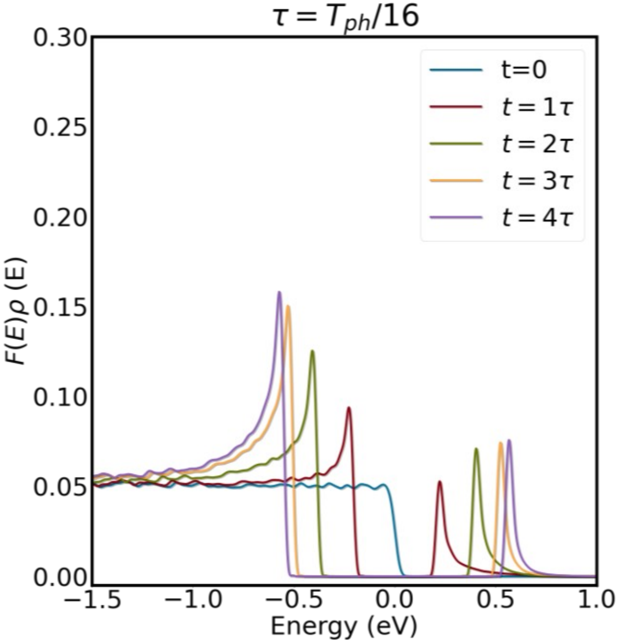
- **Optical Phonon absorption in graphene**

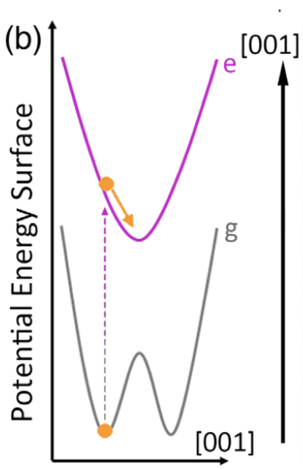


Molecular dynamics and quantum forces

- **1D chain, metal-insulator transition & electronic feedback**





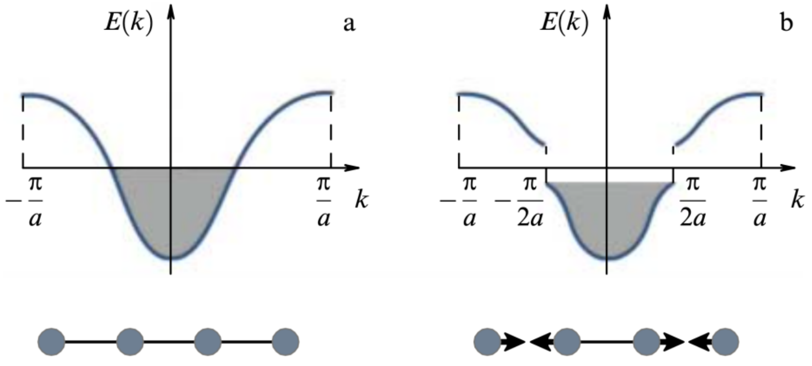


$$H(R(t)) = \sum_{\langle i,j \rangle} t(R(t)) c_i^\dagger c_j + h.c.$$

$$t(R(t)) = t_0 \exp(-3.37 \left(|R_{ij}(t)| - R_{eq} \right) / R_{eq})$$

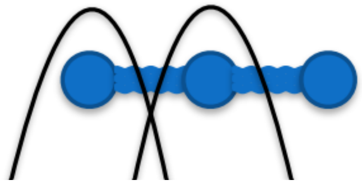
Molecular dynamics and quantum forces

- **1D chain, metal-insulator transition & electronic feedback**



$$\hat{\vec{F}}_{ij} = -N_{el} \frac{\partial H_{ij}}{\partial \vec{R}_i}$$

$$\vec{F}_i = \sum_j \frac{2t_{ij}\beta}{req} \exp\left(-\frac{\beta}{r_{eq}}\left(|\vec{r}_{ij}| - r_{eq}\right)\right) \hat{r}_{ij} Re(\langle i|j\rangle)$$



$$H(R(t)) = \sum_{\langle i,j\rangle} t(R(t)) c_i^\dagger c_j + h.c.$$

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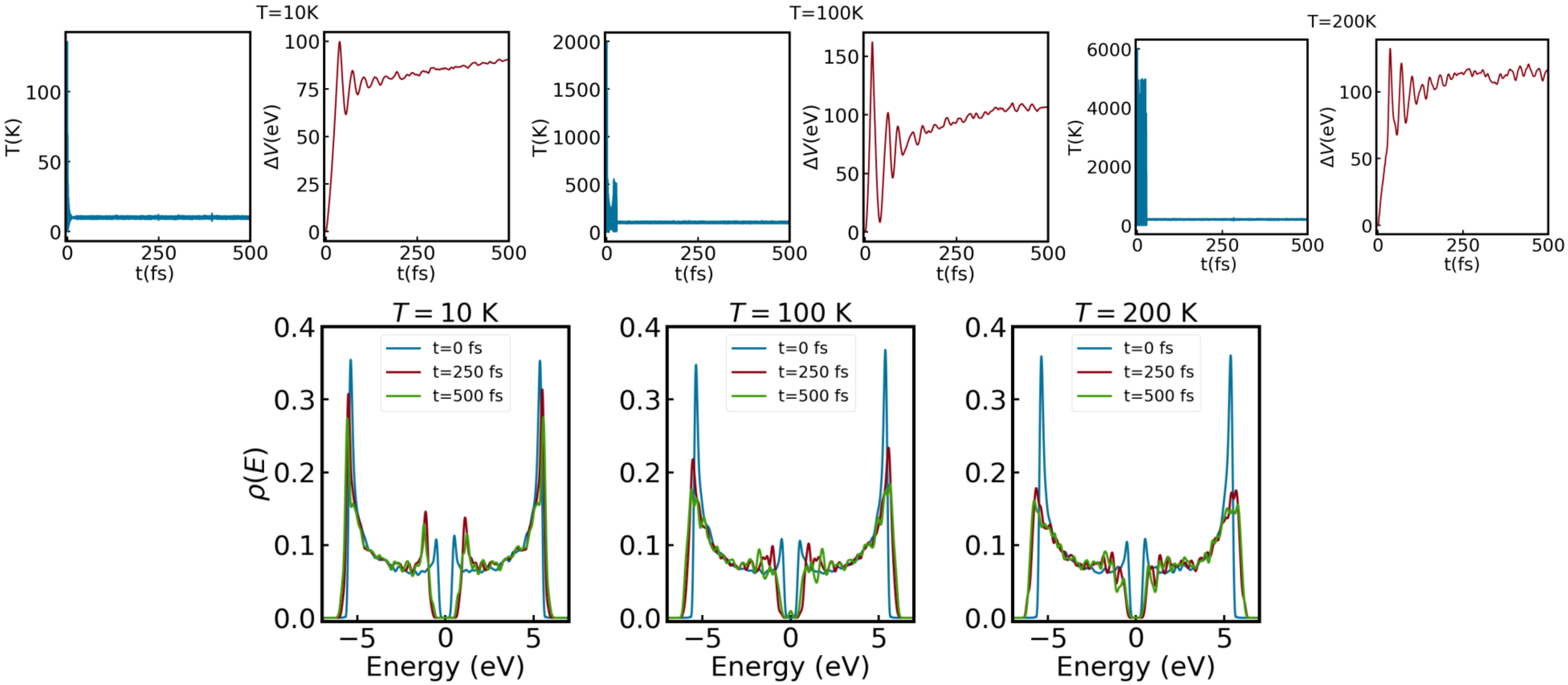
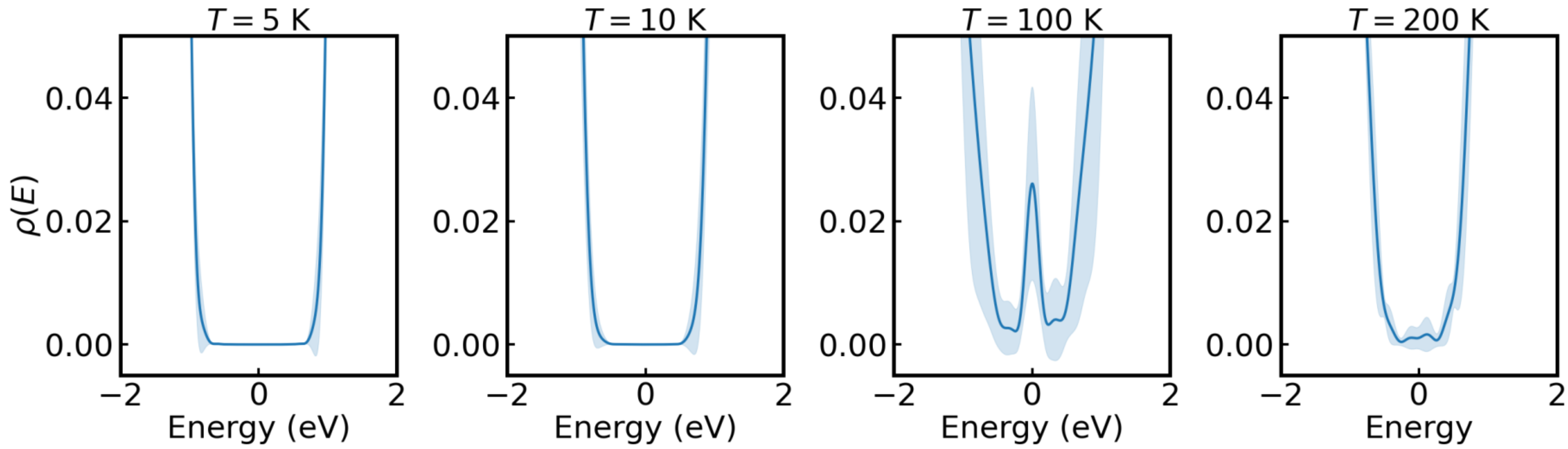
Molecular dynamics and quantum forces



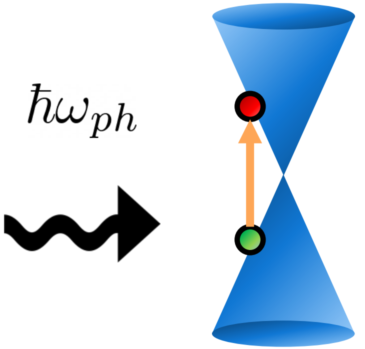
Metal-Insulator instability

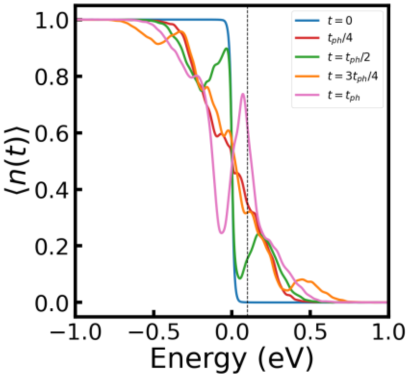


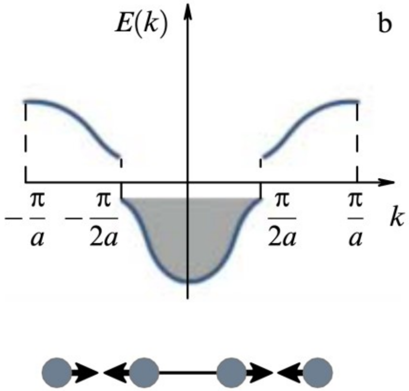
Final Considerations

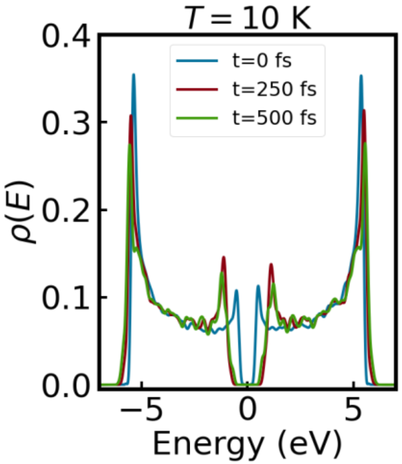
- By evaluating the time evolution of the electronic density, we can capture phonon absorption processes while maintaining the Fermi-Dirac statistics.

- By computing the quantum forces, we can include the electronic influence in the dynamics of the classical MD ions.









Thank you

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Energy Relaxation

- Phenomenological approach

$$\partial_t \rho(t) = -\frac{i}{\hbar}\left[H(t), \rho(t)\right] - i\gamma\left(\rho(t) - \rho_{eq}\right)$$

$$Tr\langle \partial_t \rho(t) \rangle = -i\gamma Tr\langle \rho(t) - \rho_{eq} \rangle$$

Energy Relaxation

- Phenomenological approach with MV operations

$$|\psi_{dyn}(t+\Delta t)\rangle = U(\Delta t)|\psi_F(t)\rangle$$

$$|\psi_F(t+\Delta t)\rangle = (1-\eta)|\psi_{Dyn}(t+\Delta t)\rangle + \eta|\psi_{Eq}(t+\Delta t)\rangle$$