Electronic correlation effects in the stopping power of ions in 2D materials

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Abstract

The energy loss of charged projectiles in correlated materials is of prime relevance for plasma-surface interaction for which we have developed a nonequilibrium Green functions (NEGF) approach. A particularly interesting effect is the *correlation induced increase* of stopping power at low velocities [1]. However, NEGF simulations are possible only for short time durations, due to the unfavorable N_t^3 scaling with the number of discretization time steps. The situation has changed radically with the recently developed G1-G2 scheme [2], which is based on the generalized Kadanoff-Baym ansatz in combination with Hartree-Fock propagators, and allows to *achieve linear scaling* with N_t. This enhancement enables us to improve previous simulations by using better selfenergies [3] and by extending the simulation duration which gives access to slower projectiles.

Stopping Power Theory

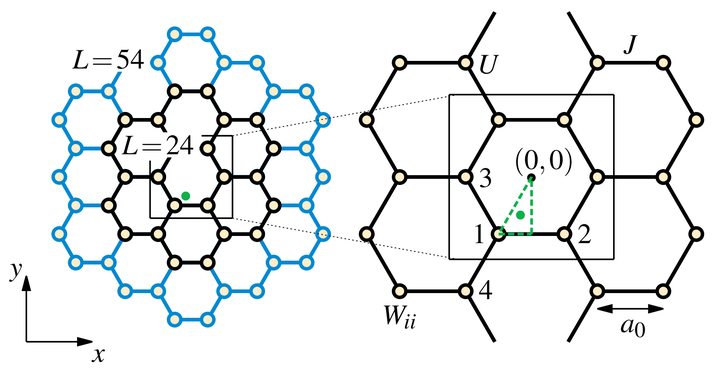

FIG. 1: 2D finite honeycomb lattice structure
L denotes the number of sites, $L = 24$ (black) $L = 54$ (blue), with the site coordinates R_i. The green point indicates the position, where the projectile hits the lattice plane. Furthermore, a_0 denotes the lattice spacing, $J(U)$ is the nearest neighbour hopping (the onsite interaction), and W_{ii} is the local energy (see below). From Ref. [4].

- The **Hubbard-Hamiltonian** of the lattice electrons

$$\hat{H}_\mathrm{e} = \hat{H}_\mathrm{hop} + \hat{H}_\mathrm{rep} + \hat{H}_\mathrm{int}$$

- **Nearest-neighbor** hopping
- On-site **Coulomb** repulsion

$$\hat{H}_\mathrm{hop} = -J \sum_{\langle i,j\rangle,\sigma} \hat{c}^\dagger_{i\sigma}\hat{c}_{j\sigma} \qquad \hat{H}_\mathrm{rep} = U \sum_i \left(\hat{n}^\uparrow_i - \frac{1}{2}\right)\left(\hat{n}^\downarrow_i - \frac{1}{2}\right)$$

- **Electron-projectile interaction**

$$\hat{H}_\mathrm{int} = \sum_{i,\sigma} W_i(t)\hat{c}^\dagger_{i\sigma}\hat{c}_{i\sigma} \quad \text{with} \quad W_i(t) = -\frac{e^2}{4\pi\epsilon_0}\frac{Z_\mathrm{p}}{|\mathbf{r}_\mathrm{p}(t) - \mathbf{R}_i|}$$

with $\hat{c}^\dagger_{i\sigma}$ ($\hat{c}_{i\sigma}$) creation (annihilation) operator on site i with spin σ and electron density $\hat{n}_{i\sigma} = \hat{c}^\dagger_{i\sigma}\hat{c}_{i\sigma}$, as well as projectile position $\mathbf{r}_\mathrm{p}$

- The bare Coulomb interaction between the projectile, the fixed background charges, and the target electrons which are initially in equilibrium is the actual cause for the stopping dynamics.
- To describe the interaction with the charged classical projectile, we use an Ehrenfest-type approach.
- Furthermore, to compute the projectiles motion, Newton´s equation with the total potential $\mathrm{V}(\mathbf{r}_\mathrm{p}, \mathrm{t})$, which includes all lattice charges, have to be solved. $Z_i(t)$ refers to the net charge on site i and m denotes the particle's mass.

$$V(\mathbf{r}_\mathrm{p}, t) = \frac{e^2}{4\pi\epsilon_0}\sum_i \frac{Z_\mathrm{p} Z_i(t)}{|\mathbf{r}_\mathrm{p}(t) - \mathbf{R}_i|} \quad \rightsquigarrow \quad m\frac{d^2}{dt^2}\mathbf{r}_\mathrm{p} = -\nabla V(\mathbf{r}_\mathrm{p}, t)$$

- The projectiles **initial position**
- Its **initial velocity**

$$\mathbf{r}_\mathrm{p} = \left(-\frac{1}{6}a_0, -\frac{\sqrt{3}}{3}a_0, -z\right) \qquad \frac{d\mathbf{r}_\mathrm{p}}{dt} = (0, 0, v_z)$$

(This position has been found to give similar stopping results compared to calculations, where one averages over many sites).

Generation of the Initial State

- The initial state is found by use of the **adiabatic switching** method:
  - Time propagation starts with noninteracting ground state
  - Interaction strength $U(t)$ is switched on adiabatically
- In the following Figures the projectile is a proton ($Z_\mathrm{p} = 1$) starting at $z = 100a_0$. Furthermore, the size of the honeycomb cluster is set to $L = 24$ sites and the nearest-neighbour hopping is a fixed value of $J = 2.8\,\mathrm{eV}$ (typical for graphene).

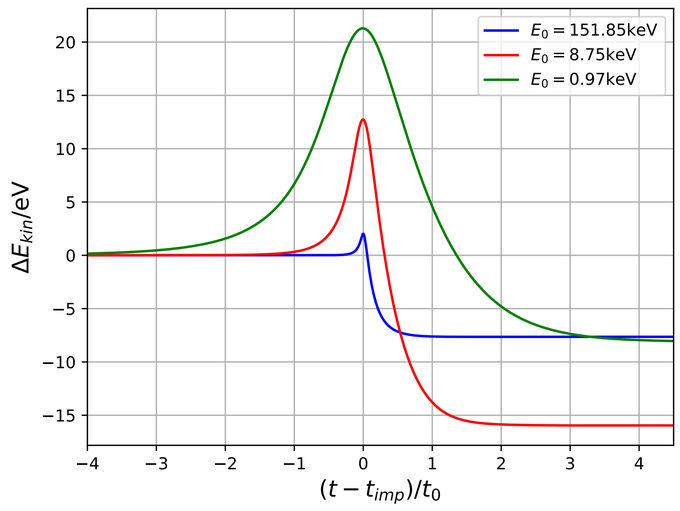

FIG. 2: Stopping dynamics
Change of the projectile's kinetic energy $\Delta E_\mathrm{kin}(t) = E_\mathrm{kin}(t) - E_\mathrm{kin}(t = 0)$ as function of time in units of $t_0 = \frac{\hbar}{J}$ for a fixed coupling strength of $\frac{U}{J} = 2$. t_imp denotes the time of the protons impact on the lattice plane. From Ref. [5].

- FIG.2 depicts three different kinetic energy curves for a slow (green), a medium-fast (red) and a fast (blue) projectile. These were created using the G1-G2 scheme with the particle-particle T-matrix selfenergy. The stopping power denotes the difference between the energy prior to and after the interaction.

G1-G2 Theory

- In general, a nonequilibrium Green functions (**NEGF**) approach was used and extended by Ehrenfest dynamics for the projectile.
- More specifically, the NEGF were used within the G1-G2 scheme, which is equivalent to the generalized Kadanoff Baym ansatz (**GKBA**).
- This scheme is particularly interesting, as for its favorable **linear time scaling**. The equations of motion using the Hartree-Fock approximation (**HF**), second-Born approximation (**SOA**) and particle-particle T-matrix (**TPP**) for the selfenergy:
  One-particle (HF):

$$i\hbar\frac{d}{dt}G^<_{ij}(t) = [h^\mathrm{HF}, G^<]_{ij}(t)$$

One-particle (SOA and TPP):

$$i\hbar\frac{d}{dt}G^<_{ij}(t) = [h^\mathrm{HF}, G^<]_{ij}(t) + [I + I^\dagger]_{ij}(t)$$

with the collision Integral $I_{ij}(t) = -i\hbar U(t)\mathcal{G}_{iiji}$ and the effective single-particle Hartree-Fock Hamiltonian h^HF (cf. Ref. [4])
Two-particle (SOA without Λ-terms):

$$i\hbar\frac{d}{dt}\mathcal{G}_{ijkl}(t) - [h^{(2),\mathrm{HF}}, \mathcal{G}]_{ijkl}(t) = \Psi_{ijkl}(t) + \Lambda^{PP}_{ijkl}(t) - [\Lambda^{PP}_{ijkl}(t)]^*$$

with $\Psi_{ijkl}(t) = (i\hbar)^2 U(t)\sum_p [G^>_{ip}(t)G^>_{jp}(t)G^<_{pk}(t)G^<_{pl}(t) - G^<_{ip}(t)G^<_{jp}(t)G^>_{pk}(t)G^>_{pl}(t)]$

and $\Lambda^{PP}_{ijkl}(t) = (i\hbar)^2 U(t) \times \sum_p [G^>_{ip}(t)G^>_{jp}(t) - G^<_{ip}(t)G^<_{jp}(t)]\mathcal{G}_{ppkl}(t)$

including $h^{(2),\mathrm{HF}}_{ijkl}(t) = \delta_{jl}h^\mathrm{HF}_{ik}(t) + \delta_{ik}h^\mathrm{HF}_{jl}(t)$

(Note that in general these equations contain spin-up/-down dependencies, but for our calculations these are irrelevant due to spin symmetry)

Stopping Power Results

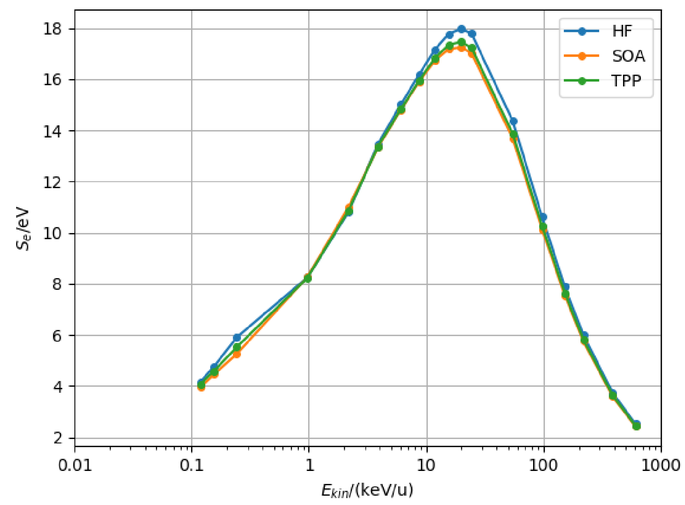

FIG. 3: Stopping power for various selfenergies
The data was generated using the G1-G2 scheme with the on-site interaction $U/J = 2$ for three different selfenergies: HF (blue), SOA (orange), TPP (green). The effect of different selfenergies is particularly strong at the maximum and at $E_\mathrm{kin} = 0.25keV/u$. From Ref. [5].

Summary

- Time-dependent simulation of ion stopping goes beyond linear response theory and includes nonlinear and non-adiabatic effects
- The electronic correlations are important for the stopping power.
- A further deviation of the selfenergies is noticeable at $E_\mathrm{kin} = 0.25keV/u$. The reason for this still has to be investigated.

Outlook

- More precise results for improved adiabatic switching
- Extension to computation of **larger system sizes**
- Improvement of the G1-G2 scheme itself by taking into account three-particle correlations.
- Simulations of even slower projectiles will provide valuable information about the extent of **correlation effects in solid targets and plasmas**
- G1-G2 scheme can be extended to plasmas using the momentum representation (see talk TO06, Thu 10:42 AM CST by C. Makait)

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