

## Problem Set 12

### Questions

- (Q.1) What is the Born–Oppenheimer approximation and how do we typically treat the electron and ion dynamics in intense laser–cluster interaction?
- (Q.2) What is the motivation for introducing so-called pseudo potentials?
- (Q.3) What is the dominating contribution to the binding energy of metal clusters: exchange or correlation?

### Problems

#### (12.1) Resonantly driven cluster

Consider a neutral, spherical jellium cluster of charge density  $\rho = \rho_i = -\rho_e$  in a laser field of amplitude  $E_0$  and frequency  $\omega$ . We know that for a static external field the electric field inside a dielectric sphere is parallel to the external field and of magnitude

$$E = \frac{3}{\epsilon_r + 2} E_0, \quad (1)$$

(Clausius-Mossotti) where  $\epsilon_r$  is the relative permittivity (or dielectric constant). Using the *Drude model* one obtains for the AC dielectric constant

$$\epsilon_r(\omega) = 1 + i \frac{\sigma(\omega)}{\epsilon_0 \omega}, \quad \sigma(\omega) = \frac{\sigma_0}{1 - i\omega/\nu}, \quad \sigma_0 = \frac{e\rho}{m\nu}$$

where  $\sigma_0$  is the DC conductivity,  $\sigma(\omega)$  is the AC conductivity, and  $\nu$  is the collision frequency. How does  $E$  in (1) behave when we replace  $\epsilon_r \rightarrow \epsilon_r(\omega)$  and the laser frequency  $\omega \gg \nu$  approaches the characteristic frequency derived in problem (11.1 iii)?

#### (12.2) Classical molecular dynamics

Let us suppose we want to simulate a cluster consisting of  $N$  singly charged ions of mass  $M$  and  $N$  electrons (mass  $m \ll M$ , charge  $-e$ ) in a laser field using classical molecular dynamics. We assume that the cluster is small compared to the laser wavelength 800 nm and that a nonrelativistic description is sufficient.

- (i) Write down the classical Hamiltonian of the system with the laser-electron interaction in (a) length form (using the electric field) and (b) in velocity form (using the vector potential). The laser-ion interaction can be safely neglected (why?).

- (ii) Write down the equations of motion for electrons and ions in both forms.
- (iii) Assume that the laser is switched on at time  $t = 0$  and switched off at time  $t_{\text{fin}}$ . Express the energy gain of the system  $\Delta\mathcal{E} = \mathcal{E}(t_{\text{fin}}) - \mathcal{E}(0)$  in terms of the work done by the laser field on the electrons in both forms.
- (iv) The singularity in the electron-ion interaction potential is smoothed,

$$V_{\text{e-ion}}(|\mathbf{R} - \mathbf{r}|) = -\frac{e^2}{|\mathbf{R} - \mathbf{r}|} \quad \longrightarrow \quad \tilde{V}_{\text{e-ion}}(|\mathbf{R} - \mathbf{r}|) = -\frac{e^2}{\sqrt{(\mathbf{R} - \mathbf{r})^2 + \epsilon^2}}.$$

The initial conditions can then be chosen such that at  $t = 0$  all particle momenta are zero, the ion positions  $\mathbf{R}_i(0) = \mathbf{R}_{0i}$ ,  $i = 1, \dots, N$  are assumed given, and the electrons sit “on top” of the ions,  $\mathbf{r}_i(0) = \mathbf{R}_{0i}$ . The smoothing parameter  $\epsilon$  can be tuned to fit the ionization potential (how?).

- (v) Assuming that the initial configuration resembles an almost spherical cluster, describe qualitatively what the laser-driven dynamics of the system will be.
- (vi) Explain why this simulation approach (a) is not appropriate for large clusters in laser fields and (b) may lead to unphysically large absorption of laser energy.