Plan of the lectures

1. Introductory remarks on metallic nanostructures
   - Relevant quantities and typical physical parameters
   - Applications
2. Linear electron response: Mie theory and generalizations
3. Nonlinear response
   - Survey of various models from N-body to macroscopic
   - Mean-field approximation (Hartree and Vlasov equations)
4. Beyond the mean-field approximation
   - Hartree-Fock equations
   - Time-dependent density functional theory (DFT) and local-density approximation (LDA)
5. Macroscopic models: quantum hydrodynamics
6. **Linear theory and comparison of various models**
7. **Illustration: the nonlinear electron dynamics in thin metal films**

Linear analysis – dispersion relations

- **Hypotheses**
  - Infinite, homogeneous system
  - Uniform ion density, $n_i = n_0$ = const.
  - Small deviations from equilibrium (“linear regime”)
  - No exchange-correlations, only Hartree and ionic potentials: $V = V_H + V_{ions}$
  - 1D
- **Wigner-Poisson equations** (equivalent to time-dependent Hartree eqs.)

$$\frac{\partial f}{\partial t} + v\frac{\partial f}{\partial x} + \frac{iem}{2\pi\hbar^2}\int\int d\lambda\, dv' e^{im(v-v')\lambda/\hbar}\left[V\left(x+\frac{\lambda}{2}\right) - V\left(x-\frac{\lambda}{2}\right)\right] f(x,v',t) = 0$$

$$\frac{\partial^2 V}{\partial x^2} = \frac{e}{\epsilon_0}\left(\int f(x,v,t)\, dv - n_0\right)$$

$$\begin{cases} f(x,v,t) = f_0(v) \\ V(x,t) = V_o = const. \end{cases}$$

Stationary solution

Linearization

$$f(x,v,t) = f_0(v) + f_1(x,v,t)$$

$$V(x,t) = \cancel{V_0} + V_1(x,t)$$

Separate equilibrium ("0") from fluctuations ("1")

$$\int f_0(v)dv = n_0$$

$$\begin{cases} \dfrac{\partial f_1}{\partial t} + v\dfrac{\partial f_1}{\partial x} + \dfrac{iem}{2\pi\hbar^2}\displaystyle\int\int d\lambda\, dv' e^{im(v-v')\lambda/\hbar}\left[V_1\left(x+\frac{\lambda}{2}\right) - V_1\left(x-\frac{\lambda}{2}\right)\right] f_0(v') = 0 \\[2em] \dfrac{\partial^2 V_1}{\partial x^2} = \dfrac{e}{\epsilon_0}\displaystyle\int f_1(x,v,t)\, dv \end{cases}$$

Linearized Wigner-Poisson eqs.

Fourier analyze in space and time (i.e., expand in plane wave basis):

$$f_1(x,v,t) \to f_1(v)\exp(-i\omega t + ikx)$$

$$V_1(x,t) \to V_1 \exp(-i\omega t + ikx)$$

In practice, this amounts to performing the following substitutions:

$$\frac{\partial}{\partial t} \to -i\omega; \qquad \frac{\partial}{\partial x} \to ik$$

We also use the identity:

$$\int \exp[im(v-v')\lambda/\hbar]\exp(\pm ik\lambda/2)d\lambda = \frac{2\pi\hbar}{m}\delta\left[v' - \left(v \pm \frac{\hbar k}{2m}\right)\right]$$

We finally obtain the Lindhard dispersion relation

$$\boxed{\underbrace{\varepsilon_L(\omega,k)}_{\text{Lindhard dielectric constant}} \equiv 1+\frac{m\omega_p^2}{kn_0}\int_{-\infty}^{+\infty}\frac{f_0(v+\hbar k/2m)-f_0(v-\hbar k/2m)}{\hbar k(\omega-kv)}\,dv = 0}$$

Using the following Taylor expansion:

$$\frac{f_0(v+\delta v)-f_0(v-\delta v)}{2\delta v} \simeq f_0'(v) + \frac{1}{6}f_0'''(v)\delta v^2 + \ldots \qquad (\text{with}: \delta v \equiv \hbar k/2m << v)$$

$$\varepsilon_L(\omega,k) \approx 1+\underbrace{\frac{\omega_p^2}{kn_0}\int\frac{f_0'(v)}{\omega-kv}\,dv}_{\text{Vlasov-Poisson dielectric const. (classical)}}+\underbrace{\hbar^2\frac{\omega_p^2 k}{24m^2n_0}\int\frac{f_0'''(v)}{\omega-kv}\,dv}_{\text{First quantum correction}}+\ldots$$

We use the following expansion, valid for small *k* (long wavelengths):

$$\frac{1}{\omega - kv} \approx \frac{1}{\omega} + \frac{kv}{\omega^2} + \frac{k^2v^2}{\omega^3} + \frac{k^3v^3}{\omega^4} + \dots$$

$$\varepsilon_L(\omega, k) \approx 1 - \frac{\omega_p^2}{\omega^2} - 3k^2\langle v^2\rangle\frac{\omega_p^2}{\omega^4} - \frac{\hbar^2k^4}{4m^2}\frac{\omega_p^2}{\omega^4} + \dots$$

This yields the following dispersion relation (“volume plasmon”):

$$\boxed{\omega^2 \approx \omega_p^2 + 3k^2\langle v^2\rangle + \frac{\hbar^2k^4}{4m^2} + \dots}$$

For a Maxwell-Boltzmann distribution:

$$\left\langle v^2 \right\rangle = v_{th}^2 = \frac{k_B T}{m}$$

For a Fermi-Dirac distribution at T=0

$$\langle v^2\rangle = \frac{\int f_0 v^2 dv}{\int f_0 dv} = \frac{v_F^2}{5} \quad \Rightarrow \quad \boxed{\omega^2 \approx \omega_p^2 + \frac{3}{5}k^2v_F^2 + \frac{\hbar^2k^4}{4m^2} + \dots}$$

Hydrodynamic dispersion relation

$$\frac{\partial n}{\partial t} + \frac{\partial (nu)}{\partial x} = 0$$

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial x} = \frac{e}{m}\frac{\partial \phi}{\partial x} + \frac{\hbar^2}{2m^2}\frac{\partial}{\partial x}\left(\frac{\partial_x^2 \sqrt{n}}{\sqrt{n}}\right) - \frac{1}{mn}\frac{\partial P}{\partial x}$$

"Classical" pressure

$$\frac{P}{P_0} = \left(\frac{n}{n_0}\right)^\gamma$$

$$P_0 = \frac{2}{5} n_0 E_F$$

Equilibrium pressure for a fermion gas at T=0

Linearization:

$$n(x,t) \simeq n_0 + n_1(x,t); \quad u(x,t) = 0 + u_1(x,t)$$

+ Fourier transform

Fluid dispersion relation

$$\boxed{\omega^2 = \omega_p^2 + \frac{\gamma}{5}k^2 v_F^2 + \frac{\hbar^2 k^4}{4m^2}}$$

Classical pressure

Quantum pressure
or Bohm potential

Wigner (Hartree) dispersion relation

$$\boxed{\omega^2 \approx \omega_p^2 + \frac{3}{5}k^2 v_F^2 + \frac{\hbar^2 k^4}{4m^2} + \dots}$$

<u>First three terms coincide if γ = 3</u>