Chap2. plasmon for Nano-particles

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§2.1 plasmon of nano-particles and Neumann-Poincaré operator

Electric permittivity of metal

This can be described by various models. A representative one is called the Drude model [Maier, 2007]. The motion of an free electron subject to an external field is governed by $m_e r''(t) + m_e \gamma r'(t) = e\vec{E}$, (2.1.1)

where m_e is the mass, e is the charge, γ is the damping constant, $\vec{E} = \vec{E}_0 e^{-i\omega t}$ is the electric field.

The displaced electron induces a dipole moment $\mu = er$, and the accumulated effect of all dipole moments from all electrons result in a macroscopic polarization $P = n\mu = ner$, where n is the number of electrons per unit volume.

Solving (2.1.1) gives $\vec{P} = -\dfrac{ne^2}{m_e(\omega^2 + i\gamma\omega)}\vec{E}$.

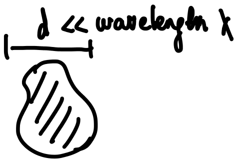
$\Rightarrow$ The electric displacement $\vec{D} = \varepsilon_0\vec{E} + \vec{P} = \varepsilon_0\left(1 - \dfrac{\omega_p^2}{\omega^2 + i\gamma\omega}\right)\vec{E}$,

where $\omega_p = \sqrt{\dfrac{ne^2}{m_e\varepsilon_0}}$ is the plasma frequency.

$\Rightarrow$ The relative permittivity of metal $\boxed{\varepsilon_m(\omega) = 1 - \dfrac{\omega_p^2}{\omega^2 + i\gamma\omega}}$. (2.1.2)

Remark: $\mathrm{Re}\,\varepsilon_m(\omega) < 0$

Mathematical models for electromagnetic scattering by nano-particles



Time-harmonic Maxwell's equations

$$\begin{cases} \nabla\times\vec{E} = i\omega\mu_0\vec{H} \\ \nabla\cdot\vec{B} = \rho \\ \nabla\times\vec{H} = -i\omega\varepsilon\vec{E} \\ \nabla\cdot\vec{B} = 0 \end{cases}$$

Typically, the size of nano-particle $d \ll$ incident wavelength λ. e.g. $d = 30\,\text{nm}$, $\lambda = 10^3\,\text{nm}$.

By scaling the problem s.t. $d = O(1)$, then the corresponding frequency $\omega \ll 1$.

Therefore, we may consider the quasi-static approximation of Maxwell's equations:

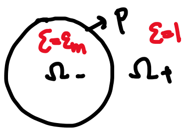
$$\begin{cases} \nabla\times\vec{E} = 0 \\ \nabla\cdot D = \rho \end{cases}$$

$\Rightarrow \exists$ electric potential u s.t. $\vec{E} = -\nabla u$. In addition u solves the Poisson equation

$$-\Delta u = \rho/\varepsilon.$$

plasmon and Neumann-poincaré operator

Consider a nano-particle placed in an static electric field $E_0 = -\nabla f$, $\Delta f = 0$. Then the electric potential u satisfies



$$\begin{cases} \Delta u = 0 & \text{in } \Omega_+ \cup \Omega_-. \\ [u] = 0 & \text{on } \Gamma \\ [\varepsilon \frac{\partial u}{\partial n}] = 0 & \text{on } \Gamma \\ u(x) - f(x) = O(\frac{1}{|x|}) & \text{as } |x| \to \infty \end{cases}$$

$$\varepsilon = \begin{cases} \varepsilon_m & \text{in } \Omega_-, \\ 1 & \text{in } \Omega_+. \end{cases}$$

Express the solution as the single layer potential

$$u(x) = \int_\Gamma \Phi(x,y)\varphi(y)\, ds_y + f(x), \quad x \in \Omega_+ \cup \Omega_-, \quad \varphi \in L_0^2(\Gamma) = \{\varphi \in L^2(\Gamma), \int_\Gamma \varphi\, ds = 0\}.$$

Taking the limit of $\frac{\partial u}{\partial n}$ to Γ yields,

$$\begin{cases} \frac{\partial u_+}{\partial n} = K'\varphi - \frac{1}{2}\varphi + \frac{\partial f}{\partial n}, \\ \frac{\partial u_-}{\partial n} = K'\varphi + \frac{1}{2}\varphi + \frac{\partial f}{\partial n}. \end{cases}$$

applying the condition $[\varepsilon \frac{\partial u}{\partial n}] = 0$ gives the integral equation

$$\frac{1}{2}\frac{1+\varepsilon_m}{1-\varepsilon_m}\varphi - K'\varphi = \frac{\partial f}{\partial n},$$ which is rewritten as $$\boxed{(\lambda - K')\varphi = \frac{\partial f}{\partial n}} \quad (2.1.3)$$

where $\boxed{\lambda = \frac{1}{2}\frac{1+\varepsilon_m}{1-\varepsilon_m}}$.

From Theorem 1.3.7, K' attains the spectral decomposition $K' = \sum_{j=1}^{\infty} \lambda_j \langle \cdot, \varphi_j \rangle_S \varphi_j$.
Therefore, we obtain the following theorem for the solution of the integral equation.

<u>Theorem 2.1.1</u> If $\lambda \neq \lambda_j$, the solution of the integral equation (2.1.3) can be expressed as $\varphi = \sum_{j=1}^{\infty} \frac{1}{\lambda - \lambda_j} \langle \frac{\partial f}{\partial n}, \varphi_j \rangle_S \varphi_j$. The electric potential

$$u(x) = \sum_{j=1}^{\infty} \frac{1}{\lambda - \lambda_j} \langle \frac{\partial f}{\partial n}, \varphi_j \rangle_S \cdot \int_\Gamma \Phi(x,y)\varphi_j(y)\, ds_y + f(x), \quad x \in \Omega_+ \cup \Omega_-.$$

<u>Excitation of plasmon</u> ε_m is given by (2.1.2).

Recall that $\lambda(\omega) = \frac{1}{2}\frac{1+\varepsilon_m(\omega)}{1-\varepsilon_m(\omega)}$. Let $\tau_j(\omega) = \lambda(\omega) - \lambda_j$. If $|\tau_j(\omega^*)| \ll 1$ for some ω^*, we call ω^* a plasmon frequency. From Theorem 2.1.1, $\|\varphi\| \gg 1$ at plasmon frequency ω^*.

Remark Note that the eigenvalues of Neumann-Poincaré operator

Remark Note that the eigenvalues of Neumann-Poincaré operator $\lambda_j \in (-\frac{1}{2}, \frac{1}{2})$ (see proposition 1.3.3). If $\mathrm{Re}\,\varepsilon_m > 0$ and $\varepsilon_m \neq 1$, then $|\lambda(\omega)| > \frac{1}{2}$ and the plasmon would not be excited. While if $\mathrm{Re}\,\varepsilon_m < 0$, then $|\lambda(\omega)| < \frac{1}{2}$, and there is possibility to excite the plasmon. Note that the latter case holds when metallic nano-particle is considered.

plasmon for more than one particle

The electric potential u satisfies

$$\begin{cases} \Delta u = 0 & \text{in } \mathbb{R}^2 \setminus (\Gamma_1 \cup \Gamma_2), \\ [u] = 0 & \text{on } \Gamma_1 \cup \Gamma_2, \\ [\varepsilon \frac{\partial u}{\partial n}] = 0 & \text{on } \Gamma_1 \cup \Gamma_2, \\ |u(x) - f(x)| = O(\frac{1}{|x|}) & \text{as } |x| \to \infty. \end{cases}$$

$$\varepsilon(x) = \begin{cases} \varepsilon_m & \text{in } \Omega_1 \cup \Omega_2, \\ 1 & \text{in } \mathbb{R}^2 \setminus (\Omega_1 \cup \Omega_2). \end{cases}$$

By expressing the solution as $u(x) = \int_{\Gamma_1} \Phi(x,y)\varphi_1(y)\,ds_y + \int_{\Gamma_2} \Phi(x,y)\varphi_2(y)\,ds_y + f(x)$, and taking the limit of $\nabla u \cdot n$ to the boundaries Γ_1 & Γ_2, it can be obtained that

$$(\lambda - \mathbb{K}')\vec{\varphi} = \begin{bmatrix} \frac{\partial f}{\partial n}\big|_{\Gamma_1} \\ \frac{\partial f}{\partial n}\big|_{\Gamma_2} \end{bmatrix}, \quad \text{where } \mathbb{K}' = \begin{bmatrix} K'_{11} & K'_{12} \\ K'_{21} & K'_{22} \end{bmatrix}, \quad \vec{\varphi} = \begin{bmatrix} \varphi_1 \\ \varphi_2 \end{bmatrix}, \text{ and}$$

$$K'_{ij}\varphi(x) = \int_{\Gamma_j} \frac{\partial \Phi(x,y)}{\partial n_x} \varphi(y)\,ds_y, \quad x \in \Gamma_i.$$

Theorem 2.1.2 The Neumann-Poincaré operator $\mathbb{K}'$ is self-adjoint in $\mathbb{H}_0^{*} := H_0^{-\frac{1}{2}}(\Gamma_1) \times H_0^{-\frac{1}{2}}(\Gamma_2)$ equipped with the inner product $\langle \vec{\varphi}, \psi \rangle_{\mathbb{S}} = \langle \mathbb{S}\vec{\varphi}, \vec{\psi} \rangle_{L^2(\Gamma_1)\times L^2(\Gamma_2)}$, where the single layer operator

$$\mathbb{S}\vec{\varphi} = \begin{bmatrix} S_{11}\varphi_1 & S_{12}\varphi_2 \\ S_{21}\varphi_1 & S_{22}\varphi_2 \end{bmatrix},$$

and $$S_{ij}\varphi(x) = \int_{\Gamma_j} \Phi(x,y)\varphi(y)\,ds_y, \quad x \in \Gamma_i.$$

The theorem can be found in [Ammari-Ciraolo-Kang-Lee-Milton, 2013].

Therefore, $\mathbb{K}'$ attains a spectral decomposition, and the resolvent $(\lambda - \mathbb{K}')$ can be obtained.