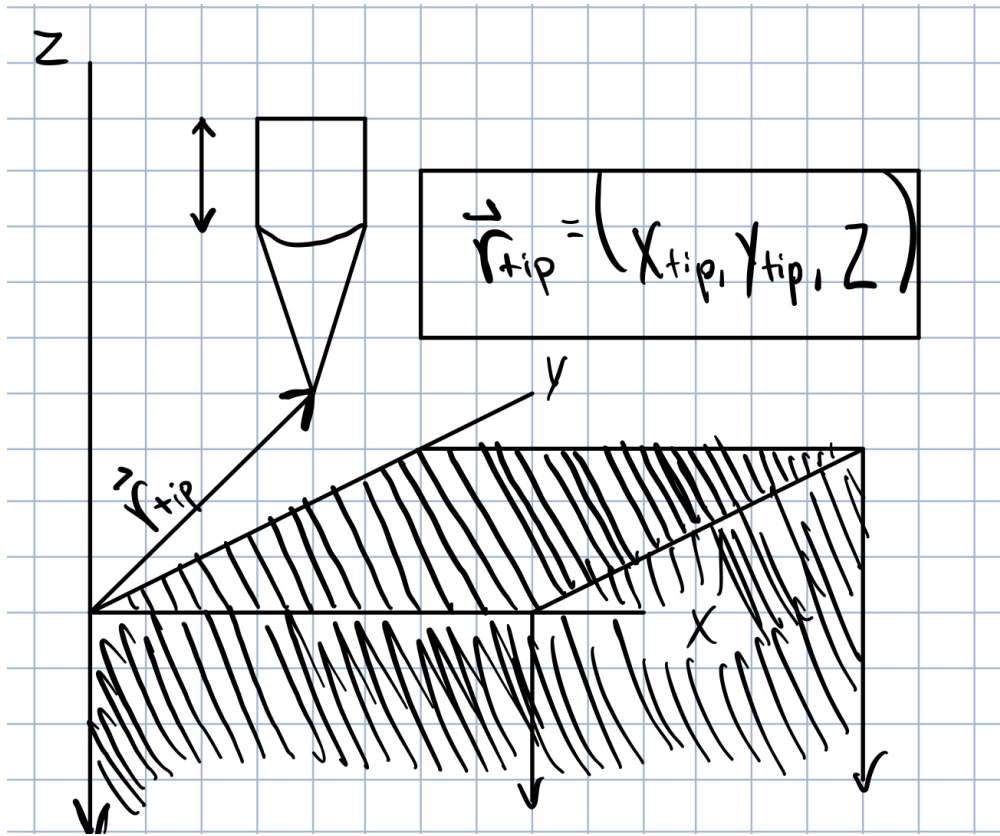


MODELING PLASMONS AND SNOM RESPONSE

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1. THEORETICAL FRONT MATTER

Assume we have a system consisting of an azimuthally symmetric tip and a sample modeled as an infinite half-plane.



We begin by defining the eigenmodes of these two individual systems (tip and sample) which we will then use to model the response of the two elements in the system to each other.

1.1. Tip Eigenmodes.

[TO BE UPDATED]

The generalized spectral method affords a solution to the electrostatic scattering problem comprising an illuminated object (e.g. a near-field probe) interacting with a dielectric half space. In this approach, the dipole moment of the tip can be computed as [insert reference

to Bor-Yuan & Misha’s APL paper]:

$$p_z = \sum_{\alpha} \frac{\Lambda_{\alpha}^z}{\beta_{\alpha}^z - \beta} \quad (1.1)$$

This expression expands the probe-sample interaction according to a discrete infinite series of "eigenfields" (also denoted "gap polaritons") satisfying all boundary conditions of the electrostatic problem, each self-sustaining (without incident field) in the fictitious case where $\beta = \beta_{\alpha}^z$. Here β_{α}^z (determined in part by probe-sample distance z) are complex numbers constrained in fact to the lower half complex plane. The numbers Λ_{α}^z pertain to the charge distribution induced on the probe associated with these gap polaritons, and determine in part the total probe polarization.

Although gap-propagation and probe polarization both uniformly preserve the structure of gap polaritons, the case of a more complex sample geometry implies inter-mode scattering at the sample surface. We may describe this possibility by a generalized scattering matrix R expressed in the eigenmode basis, whereby the probe polarization becomes

$$p_z = \sum_{\alpha} (\beta^z - R)_{\alpha\beta}^{-1} \Lambda_{\beta}^z \quad (1.2)$$

Here β_{α}^z is a diagonal matrix of the gap eigenvalues β_{α}^z .

In particular, this scattering matrix R :

- (1) Defines how the sample scatters a field from the tip
- (2) Projects this scattered response back onto the set of gap polaritons. For simplicity, we preserve only those gap eigenmodes with azimuthal symmetry about the origin coordinate \mathbf{r}_{tip} . Qualitatively, this approximation selects only the gap polaritons with appreciable ability to induce net polarization on the probe, while discarding others; implications of this approximation are not yet entirely clear.

Notationally, we first define the tip eigenmodes (pairs of corresponding gap eigenvalues and eigenfunctions) as:

$$\mathbb{T} = \{\beta_{\alpha}^z, \varphi_{\alpha}^z\} \quad (1.3)$$

where \mathbb{T} is the set of gap eigenpairs, β_{α}^z is the eigenvalue at index α and φ_{α}^z is the corresponding eigenfunction. Whenever we refer to a gap eigenmode, we will use Greek symbols. Note that these eigenmodes are also indexed by z as they change with the distance z between tip and sample.

At the moment, we model the gap eigenfunctions in our code as a finite set of zeroth order Bessel functions. We would like extend this representation later [any simple representation we can use for now for testing purposes?].

1.2. Sample response.

We first consider the electrodynamic response of the 2-dimensional sample, which produces an induced electrostatic potential $\Phi_{\text{ind}}(\vec{r})$ in response to an external potential $\Phi_{\text{ext}}(\vec{r})$, implying that the total potential is given by $\Phi(\vec{r}) = \Phi_{\text{ext}}(\vec{r}) + \Phi_{\text{ind}}(\vec{r})$. The induced potential is associated with "free charges" induced on the sample as dictated by the 2-dimensional conductivity σ and the continuity of charge driven to oscillate at excitation frequency ω :

$$-i\omega\rho + \nabla \cdot \mathbf{j} \implies \rho = \nabla \cdot \left[\frac{i\sigma}{\omega} \cdot -\nabla\Phi(\vec{r}) \right] \quad (1.4)$$

$$\therefore \Phi_{\text{ind}}(\vec{r}) = \Phi(\vec{r}) - \Phi_{\text{ext}}(\vec{r}) = V * \nabla \cdot \left[-\frac{i\sigma}{\omega} \nabla\Phi(\vec{r}) \right] \quad (1.5)$$

Here V is a (energy- and position-dependent) Coulomb kernel operator whose operation in free-space is given for example by:

$$V * f(\vec{r}) \equiv \int_{\Omega} d^2\vec{r}' \frac{1}{|\vec{r} - \vec{r}'|} f(\vec{r}'). \quad (1.6)$$

The product $q_p(\vec{r}) \equiv i\omega/\sigma(\vec{r})$ establishes a local length scale for the electrostatic response of the sample. In the case of a well-conductive sample, the complex-valued conductivity $\sigma \equiv \sigma_1 + i\sigma_2$ will be dominated by a (positive) imaginary part. Then we can define a real-valued dimensionless field $Q(\vec{r})$ to describe spatial variations of the conductivity about its mean value, a “typical” inverse length scale q_ω (real part of the characteristic plasmon wave vector at energy ω), and a complex-valued constant $\tilde{\alpha}_\sigma$ related via:

$$q_p(\vec{r}, \omega) \equiv \tilde{\alpha}_q^{-1} Q_p(\vec{r}, \omega) q_\omega \equiv \tilde{\alpha}_q^{-1} Q_p(\vec{r}, \omega) \frac{\omega}{\langle |\sigma(\vec{r})| \rangle}. \quad (1.7)$$

Here $\langle |\sigma(\vec{r})| \rangle$ denotes the spatial average absolute value of the conductivity, and $\tilde{\alpha} \equiv \langle -i\sigma/|\sigma| \rangle$ is a unit-norm constant with a (small) negative imaginary part when $\sigma_1 > 0$.

Now Eq. 1.5 establishes a self-consistent integro-differential equation for Φ expressible compactly as:

$$\begin{aligned} \tilde{\alpha}_\sigma \hat{V} \hat{L} \Phi(\vec{r}) - q_\omega \Phi(\vec{r}) &= -q_\omega \Phi_{\text{ext}}(\vec{r}) \\ \text{with } \hat{L} f(\vec{r}) &\equiv \nabla \cdot [Q_p(\vec{r})^{-1} \nabla f(\vec{r})] \\ \text{and } \hat{V} f(\vec{r}) &\equiv \int_{\Omega} d^2\vec{r}' V(\vec{r} - \vec{r}') f(\vec{r}'). \end{aligned} \quad (1.8)$$

1.3. Sample Eigenmodes.

Eq. 1.8 can be solved in terms of eigenmodes of the operator \hat{L} , defined through a homogeneous Helmholtz equation:

$$(\hat{L} - q_n^2)u_n = 0 \quad (1.9)$$

from which we obtain eigenmodes:

$$\mathbb{S} = \{q_n, u_n\} \quad (1.10)$$

where \mathbb{S} is the set of eigenpairs of \hat{L} , q_n is the eigenvalue indexed by n , and u_n is the corresponding eigenfunction. This defines a set of orthonormal basis vectors $\{|u_n\rangle\}$ for the space of induced electrostatic potential functions satisfying the appropriate boundary conditions on the sample.

Related to this eigenvalue problem is the homogeneous plasmon equation for our system with a constant but complex-valued conductivity σ :

$$(\hat{V}\hat{L} - i\omega/\sigma)\Phi = 0 \quad (1.11)$$

This homogeneous problem is an initial value problem solved by a linear combination of eigenmodes decaying in time with complex frequencies ω_n . In the case of an entirely screened (local) Coulomb interaction $\hat{V} \equiv v$, these frequencies are given by $\omega_n = -i\sigma v q_n^2$. Going further, in the case of a Drude conductivity $\sigma \propto il/\omega$ with l some intrinsic length scale, here we establish the acoustic plasmon dispersion: $\omega_n \propto \sqrt{lv} q_n$. On the other hand, in the case of the unscreened Coulomb kernel Eq. 1.6 where we might approximate $\hat{V}u_n \sim q_n^{-1}u_n$, we obtain the conventional dispersion for unscreened plasmons in a 2-dimensional electron gas: $\omega_n \propto \sqrt{l} q_n$.

By adding a source term $f = -q_\omega \cdot \Phi_{\text{ext}}$ to our plasmon equation, we obtain the inhomogeneous Helmholtz equation for our system:

$$(\tilde{\alpha}_\sigma \hat{V}\hat{L} - q_\omega)\Phi = f. \quad (1.12)$$

We can represent Eq. 1.12 in matrix form by projecting our functions and operators onto the eigenbasis of \hat{L} , whereby $|\Phi\rangle = \sum_n \Phi_n |u_n\rangle$ and $|f\rangle = \sum_n \langle u_n|f\rangle |u_n\rangle$ and:

$$\hat{L} = \sum_n q_n |u_n\rangle \langle u_n| \quad (1.13)$$

$$\hat{V} = \sum_{n,m} |u_m\rangle \langle u_m|V|u_n\rangle \langle u_n|. \quad (1.14)$$

If we consider $V_{n,m} \equiv \langle u_m|V|u_n\rangle$ matrix elements for the Coulomb operator \mathbf{V} , we obtain a solution for Φ in the form of a matrix equation:

$$\vec{\Phi} = (\tilde{\alpha}_\sigma \mathbf{Q} \mathbf{V} - q_\omega \mathbf{I})^{-1} \vec{f} \quad (1.15)$$

$$= \frac{q_\omega}{q_\omega - \tilde{\alpha}_\sigma \mathbf{Q} \mathbf{V}} \vec{\Phi}_{\text{ext}} \quad (1.16)$$

$$\equiv \mathbf{S}_{\sigma,\omega} \vec{\Phi}_{\text{ext}} \quad (1.17)$$

Here \mathbf{I} is the identity matrix, \mathbf{Q} is the diagonal matrix of squared eigenvalues such that $Q_{n,n} \equiv q_n^2$, and $\mathbf{S}_{\sigma,\omega}$ defines a scattering matrix for the sample.

In the case that \mathbf{V} is also a diagonal matrix (e.g. a local Coulomb interaction, and not necessarily meaningful for our physics of interest) with elements v_n , then Eq. 1.16 becomes:

$$\Phi(\vec{r}) = \int_\Omega G(\vec{r}, \vec{r}') \Phi_{\text{ext}}(\vec{r}') \quad (1.18)$$

$$\text{with } G(\vec{r}, \vec{r}') \equiv \sum_n q_\omega \frac{u_n(\vec{r}) u_n^*(\vec{r}')}{q_\omega - \tilde{\alpha}_\sigma v_n q_n^2} \quad (1.19)$$

Here G is the familiar Green's function associated with the operator product $\tilde{\alpha}_\sigma \hat{V} \hat{L}$. Unfortunately, our life will not generally be so easy as 1.19, and we will have to use Eq. 1.16.

1.4. Basis Changing.

It is generally necessary to project from the gap eigenmode basis \mathbb{T} to the sample eigenmodes basis \mathbb{S} , and vice versa. It will therefore be useful to have a change of basis matrix $U_{n\alpha}$ for this purpose.

$$U_{n\alpha} = \langle u_n | \varphi_\alpha \rangle = \int d^3r u_n^*(\mathbf{r}) \varphi_\alpha(\mathbf{r}) \quad (1.20)$$

$$U_{\alpha n} = U_{n\alpha}^{-1} \quad (1.21)$$

For non-square representations of U , here we imply the generalized pseudo-inverse.

To obtain our scattering R matrix in the gap eigenbasis:

$$R_{\alpha\beta} = U_{\alpha n}^{-1} S_{nm} U_{m\beta}^{-1} \quad (1.22)$$

where S_{nm} denotes elements of the sample scattering matrix $\mathbf{S}_{\sigma,\omega}$ identified in Eq. 1.17.

2. OUR JOB: COMPUTING THIS STUFF

Generally stated, our job is to compute what potential our tip is going to see at a given point $\mathbf{r}_{\text{tip}} = (x_{\text{tip}}, y_{\text{tip}})$.

The steps for this computation are given below in Pythonic pseudocode

1. Find sample eigenmode basis `u[n]`
2. Find `V[n,m]`, `R[n,m]`
3. for all `r_tip`:
4. for `z` in `tip_vibration_period`:
5. Find tip eigenmode basis `phi[alpha, r_tip, z]`
6. Calculate basis transformation `U[n, alpha]` (equation 1.13)
7. Calculate `R[alpha, beta]` (equation 1.15)
8. Calculate `P[alpha, z]` (equation 1.3)
9. Calculate `p[z]` (equation 1.2)
10. Sum `p[z]`