Multiple-Scattering Formalism Beyond the Quasistatic Approximation

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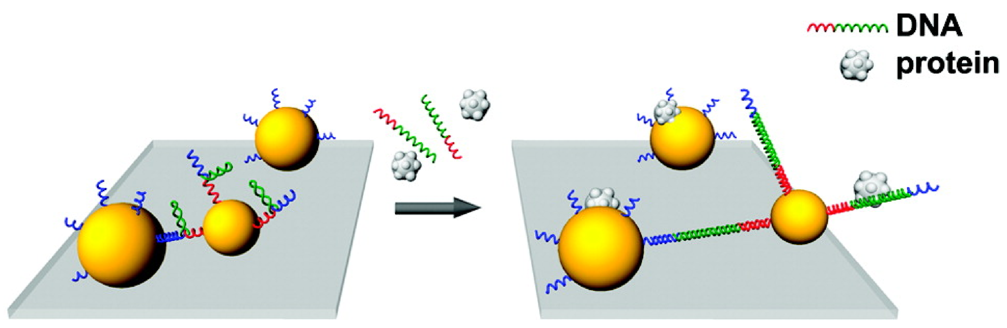
I: What Applications of Formalism?

- Modeling of optical microstructures, e.g. 3D photonic crystals; Computation of Green's tensor, LDOS, cavity modes and Q-factors.
- Modeling of plasmonic nanoparticles, for instance for use in biosensing [1] and imaging and detection devices.

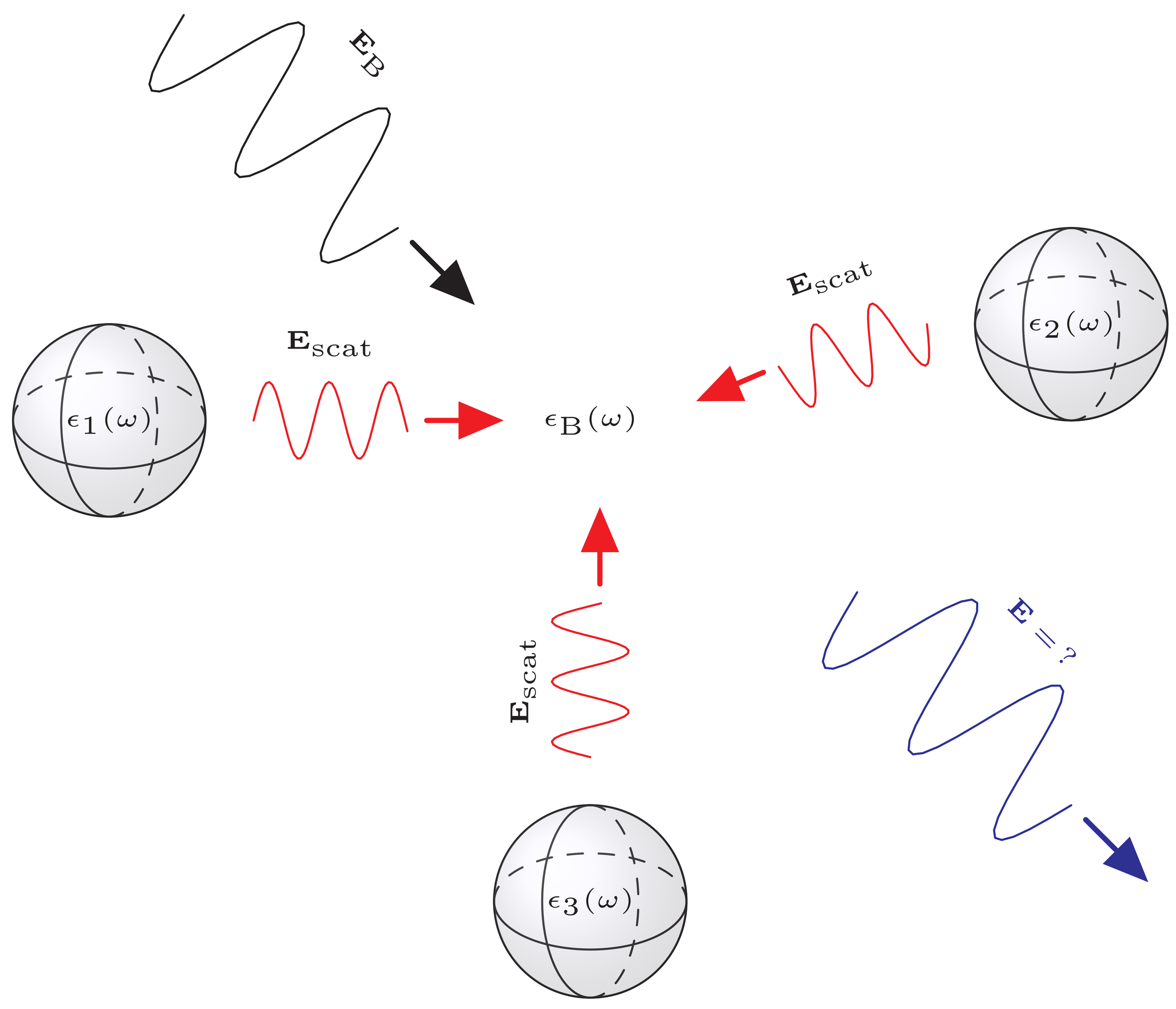


II: Why Multiple-Scattering Formalism?

- Intriguing phenomena, e.g. near-field coupling and strong field enhancements, for closely spaced metallic nanoparticles.
- Breakdown of simpler approaches, e.g. quasistatic and dipole approximations, for closely spaced particles; Need for modeling beyond lowest-order description.

III: Problem Formulation

- Rigorous determination of $\mathbf{E}$, resulting from scattering of $\mathbf{E}_\mathrm{B}$ on N spherical scatterers in homogeneous background



- Computation of derived quantities, e.g. far-field radiation pattern, $\mathbf{f}(\theta,\phi)$, extinction cross section, C_ext, and LDOS, $\rho(\mathbf{r};\omega)$.

IV: Solution Technique

- Lippmann-Schwinger equation

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_\mathrm{B}(\mathbf{r}) + k_0^2 \int_{V_\mathrm{scat}} \mathbf{G}_\mathrm{B}(\mathbf{r},\mathbf{r}')\Delta\epsilon(\mathbf{r}')\mathbf{E}(\mathbf{r}')\,\mathrm{d}\mathbf{r}'.$$

- Implicit equation inside scatterers; Explicit outside scatterers.
- Explicit error estimate.
- 3D generalization of [2]; Local expansion of field inside scatterers on spherical wavefunctions

$$\mathbf{E}(\mathbf{r}_j) = \sum_{\alpha\, l\, m} a_{j\,\alpha\, l\, m}\psi^j_{l,m}(\mathbf{r}_j)\,\mathbf{e}_\alpha, \quad \psi^j_{l,m}(\mathbf{r}_j) = K^l_j j_l(k_j r_j) Y^m_l(\theta_j,\phi_j).$$

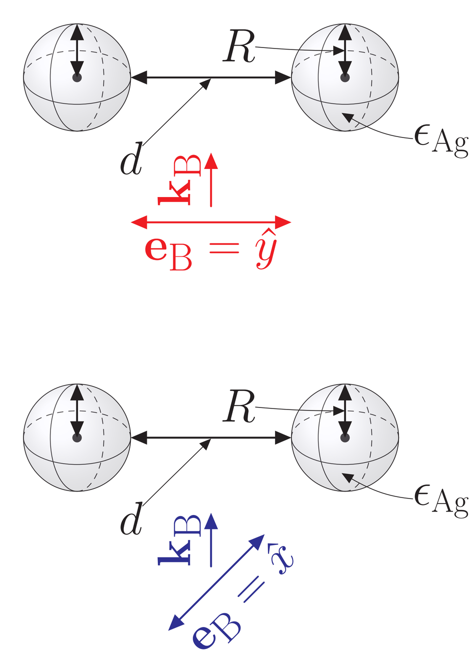
- Lippmann-Schwinger equation as matrix equation for expansion coefficients

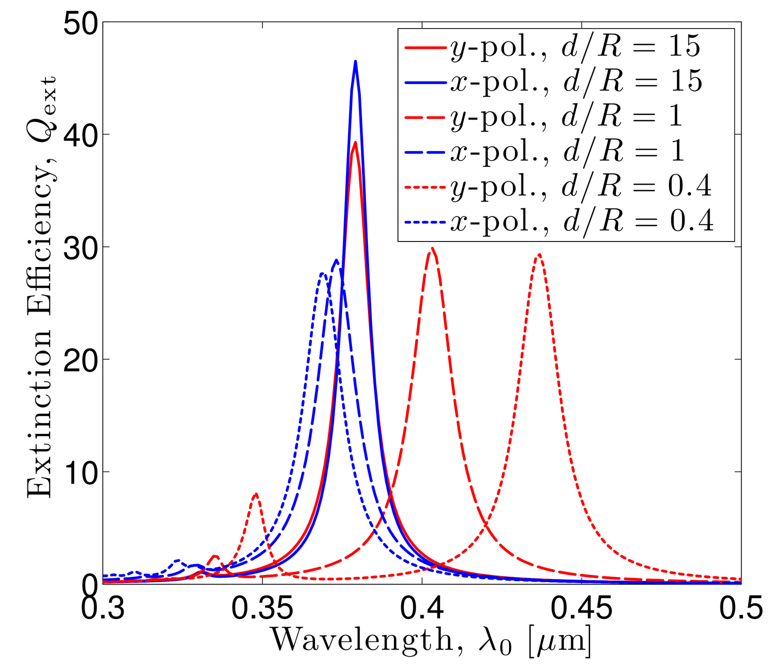
$$\mathbf{a} = \mathbf{M}\mathbf{a}_\mathrm{B} + k_0^2\mathbf{G}\boldsymbol{\Delta\epsilon}\mathbf{a}.$$

- All matrix elements expressed analytically.

V: Plasmonic Dimer

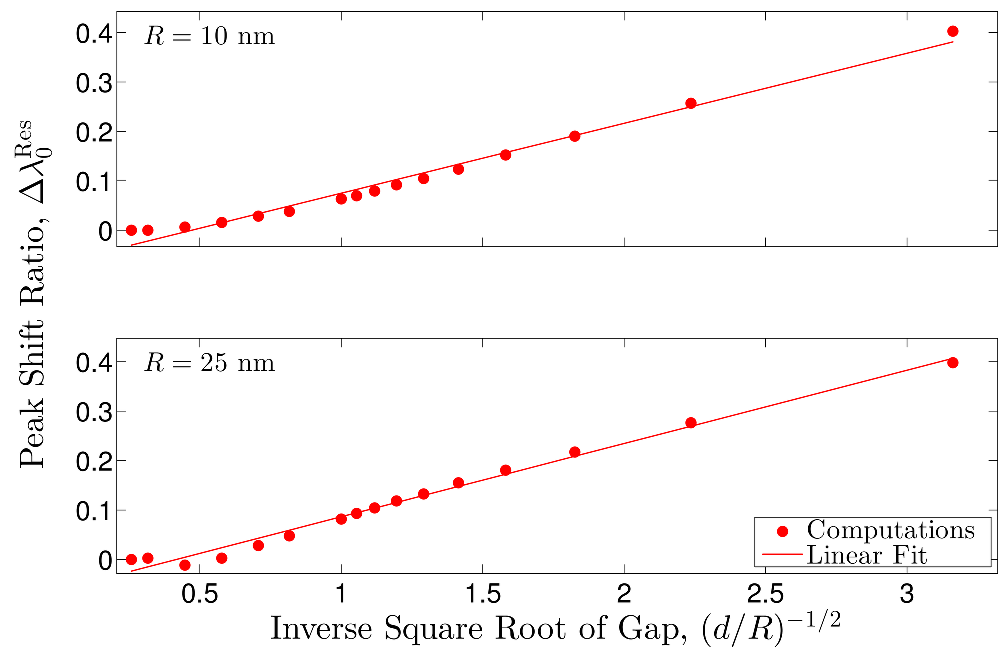
- Plane wave excitation; Parallel ($\hat{y}$) and perpendicular ($\hat{x}$) polarizations





- Distinct redshift for decreasing d/R for parallel polarization.
- Correlate peak wavelength with d/R through peak shift ratio

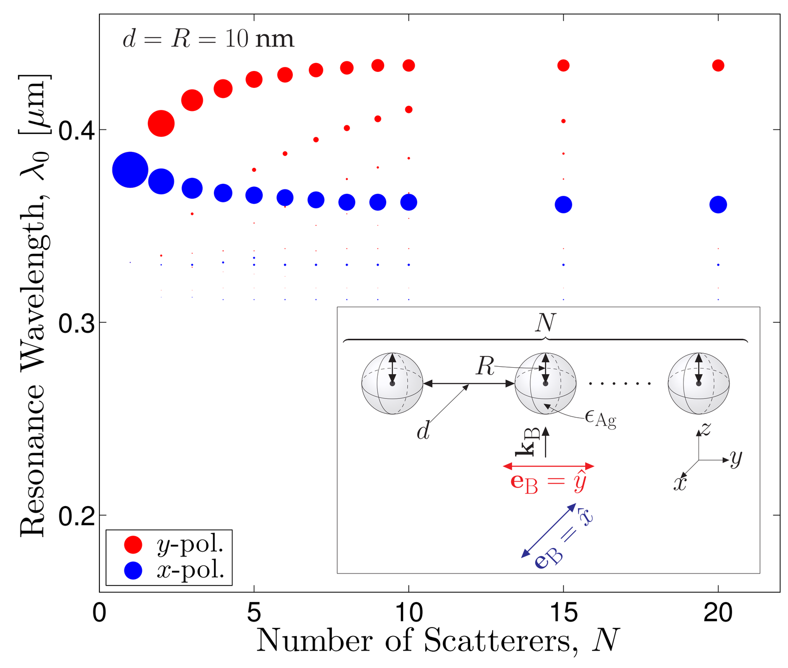
$$\Delta\lambda_0^\mathrm{Res} \equiv \frac{\lambda_0^\mathrm{D} - \lambda_0^\mathrm{D,single}}{\lambda_0^\mathrm{D,single}},$$



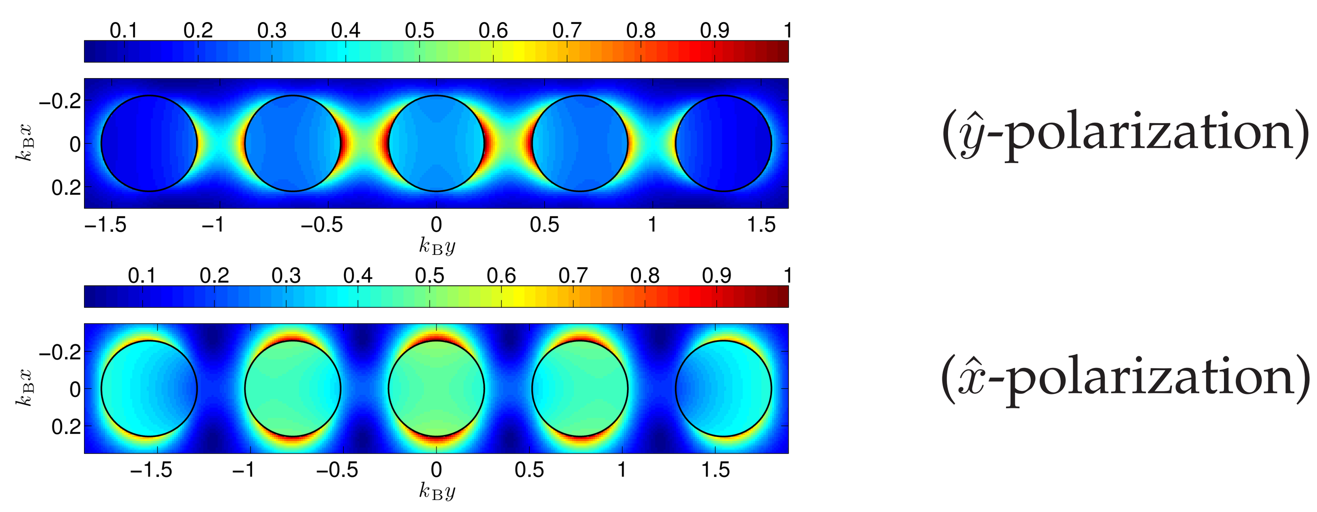
- $\Delta\lambda_0^\mathrm{Res} \propto \exp(-(d/R)/\eta)$ [3]; $\Delta\lambda_0^\mathrm{Res} \propto 1/(d/R)$ [4]; Not agreement.

VI: Plasmonic Chain

- Resonance wavelengths as function of number of scatterers



- $\Delta\lambda_0^\mathrm{Res} = \Delta\lambda_0^\mathrm{Asymp}\exp(-\eta_N/(N-1))$, $\eta_N \simeq 2$; Agreement with [4].
- Two polarizations; Two types of localized surface plasmons



VII: Conclusions

- Multiple-scattering formalism based on Green's tensor; Easy and intuitive access to, e.g., extinction scattering spectra, LDOS and cavity modes/quasinormal modes.
- Plasmonic dimer: Correlation between spectrum and particle spacing ($\hat{y}$-pol.).
- Plasmonic chain: Finite interaction length along chain ($\hat{y}$-pol.).

References

[1] J. I. L. Chen, Y. Chen, and D. S. Ginger, J. Am. Chem. Soc. **132**, 9600 (2010).

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[3] P. K. Jain, W. Huang, and M. A. El-Sayed, Nano Lett. **7**, 2080 (2007).

[4] N. Harris, M. D. Arnold, M. G. Blaber, and M. J. Ford, J. Phys. Chem. C **113**, 2784 (2009).