Sub-wavelength resonances: From super-resolution to metamaterials

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Concept of super-resolution

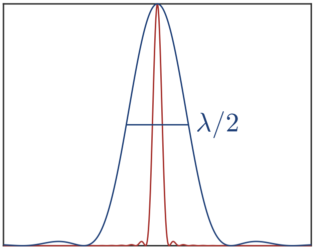
- Resolution: smallest detail that can be resolved.
- G_{k_m}: outgoing fundamental solution to $\Delta + k_m^2$; $k_m := \omega/\sqrt{\varepsilon_m}$.
- Helmholtz-Kirchhoff identity $\Rightarrow$ Resolution: determined by the behavior of the imaginary part of the Green function.

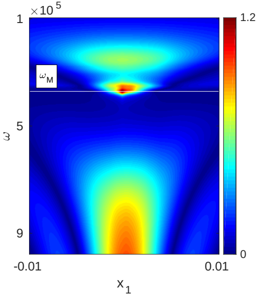
$$\Im m\, G_{k_m}(x, x_0) = k_m \int_{|y|=R} \overline{G_{k_m}(y, x_0)} G_{k_m}(x, y) ds(y), \quad R \to +\infty.$$

- $\min_x \int_{|y|=R} |G_{k_m}(x, y) - G_{k_m}(y, x_0)|^2 ds(y)$.
- $\Im m\, G_{k_m}$: point spread function.
- The more point-like $\Im m\, G_{k_m}$ is, the sharper the resolution.

Concept of super-resolution

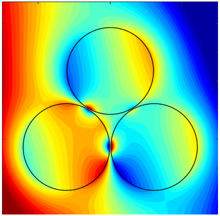
- Reduce the focal spot size;
- Confine waves to length scales significantly smaller than half the wavelength.





Sub-wavelength wave physics

- Nanophotonics and nanophononics:
  - Focus, control, manipulate, reshape, guide waves at sub-wavelength scales.
  - Mathematical and numerical framework for sub-wavelength wave physics that explains quantitatively the mechanisms behind the spectacular properties exhibited by metamaterials in recent physical experiments.

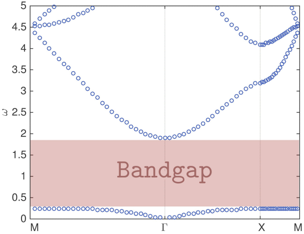


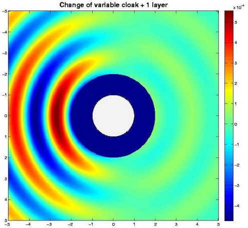
Sub-wavelength resonances

- Sub-wavelength resonators: size $< 2\pi/$ resonant frequency
  - Helmholtz resonators;
  - plasmonic nanoparticles;
  - Minnaert bubbles.
- Microstructured resonant media.
- Building block microstructure: sub-wavelength resonator.

Sub-wavelength resonances

- Super-focusing of waves;
- Negative material parameters: invisibility and cloaking; super-lensing;
- Metasurfaces: planar structures that locally modify the polarization, phase and amplitude of light or sound in reflection or transmission;
- Sub-wavelength band gap materials: microstructure periodicity smaller than the wavelength; prohibited low-frequency wave propagation.
- Unify the mathematical theory of super-resolution, sub-wavelength bandgap materials, metamaterials, and cloaking.





Sub-wavelength resonances

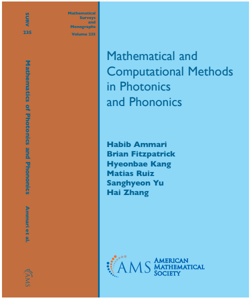
- Microstructured resonant media:
    - Dilute regime (Small-volume fraction of the sub-wavelength resonators): Effective medium theory:
        - High contrast materials: slightly below the free space resonant frequency.
        - Super-resolution and super-focusing of waves.
        - Negative effective refractive index $\Rightarrow$ Sub-wavelength bandgap opening slightly above the free space resonant frequency.
    - Non-dilute regime:
        - High-frequency homogenization techniques.
        - Super-focusing slightly below a critical frequency.
        - Sub-wavelength bandgap opening slightly above a critical resonance.
        - Critical frequency $\neq$ free space resonant frequency.

Sub-wavelength resonances

- Hybridization for arbitrary number of strongly interacting sub-wavelength resonators:
  - Singular hybridization method.
  - Double-negative materials.
  - Broadband metamaterials.

Super-resolution in resonant media

- Helmholtz resonators: Work with H. Zhang (Comm. Math. Phys., 2015);
- Plasmonic nanoparticles: Works with P. Millien, M. Ruiz, S. Yu, H. Zhang (Arch. Ration. Mech. Anal., 2016, J. Diff. Equat., 2016, Proc. Royal Soc., 2015, SIAM Rev., 2018);
- Minnaert bubbles: Works with B. Fitzpatrick, D. Gontier, H. Lee, S. Yu, H. Zhang (SIAM J. Math. Anal. 2017, SIAM J. Appl. Math. 2017, J. Diff. Equat. 2017, Proc. Royal Soc. A, 2017, Ann. IHP C);
- Lecture Notes (with B. Fitzpatrick, H. Kang, M. Ruiz, S. Yu, H. Zhang).



Super-resolution in resonant media

- The sharper is $\Im m\, G_{k_m}$, the better is the resolution.
- Local resonant media used to make sharp peaks of $\Im m\, G_{k_m}$.
- Mechanism of super-resolution in resonant media[1] :
  - Interaction of the point source x_0 with the resonant structure excites high-modes.
  - Resonant modes encode the information about the point source and can propagate into the far-field.
  - Super-resolution: only limited by the resonant structure and the signal-to-noise ratio in the data.

[1] with H. Zhang, Proc. Royal Soc. A, 2015.

Super-resolution in resonant media

- System of weakly coupled sub-wavelength resonators.
- Size of the resonator $\delta \ll$ wavelength $2\pi/k_m$; distance between the resonators of order the resonator's size.
- $\Im m\, G^\delta = \Im m\, G_{k_m} +$ exhibits sub-wavelength peak with width of order one $\Rightarrow$ Break the resolution limit.

Super-resolution in resonant media

- Sub-wavelength resonators

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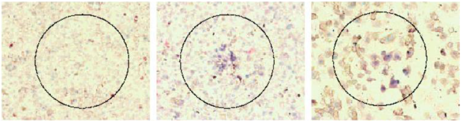
- Asymptotic expansion of the Green function[2] (δ: size of the resonator openings; z_j: center of aperture for jth resonator; J: number of resonators; $\omega = O(\sqrt{\delta})$):

$$\Im m\, G^{\delta}(x, x_0, \omega) \approx \frac{\sin \omega |x - x_0|}{4\pi |x - x_0|} + \sqrt{\delta} \sum_{j=1}^{J} \frac{c_j}{|x - z_j|\,|x_0 - z_j|}.$$

[2] with H. Zhang, Comm. Math. Phys., 2015.

Plasmonic resonances for nanoparticles

- Gold nano-particles: accumulate selectively in tumor cells; bio-compatible; reduced toxicity.
- Detection: localized enhancement in radiation dose (strong scattering).
- Ablation: localized damage (strong absorption).
- Functionalization: targeted drugs.



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Plasmonic resonances for nanoparticles

- D: nanoparticle in $\mathbb{R}^d, d = 2, 3$; $\mathcal{C}^{1,\alpha}$ boundary ∂D, $\alpha > 0$.
- $\varepsilon_c(\omega)$: complex permittivity of D; $\varepsilon_m > 0$: permittivity of the background medium;
- Permittivity contrast: $\lambda(\omega) = (\varepsilon_c(\omega) + \varepsilon_m)/(2(\varepsilon_c(\omega) - \varepsilon_m))$.
- Causality $\Rightarrow$ Kramer-Krönig relations (Hilbert transform); Drude model for the dielectric permittivity $\varepsilon_c(\omega) = 1 - \omega_p^2/\omega^2$; $\varepsilon_m = 1$.
- G: Fundamental solution to the Laplacian; $\mathcal{S}_D$: Single-layer potential.
- Neumann-Poincaré operator $\mathcal{K}_D^*$:

$$\mathcal{K}_D^*[\varphi](x) := \int_{\partial D} \frac{\partial G}{\partial \nu(x)}(x - y)\varphi(y)\, ds(y)\ , \quad x \in \partial D.$$

  ν: normal to ∂D.
- $\mathcal{K}_D^*$: compact operator on $L^2(\partial D)$; Spectrum of $\mathcal{K}_D^*$ lies in $]-\frac{1}{2}, \frac{1}{2}]$ (Kellog).

Plasmonic resonances for nanoparticles

- $\mathcal{K}_D^*$ self-adjoint on $L^2(\partial D)$ iff D is a disk or a ball.
- Symmetrization technique for Neumann-Poincaré operator $\mathcal{K}_D^*$:
  - Calderón's identity: $\mathcal{K}_D \mathcal{S}_D = \mathcal{S}_D \mathcal{K}_D^*$;
  - In three dimensions, $\mathcal{K}_D^*$: self-adjoint in the Hilbert space $\mathcal{H}^*(\partial D) = H^{-\frac{1}{2}}(\partial D)$ equipped with

$$(u, v)_{\mathcal{H}^*} = -(u, \mathcal{S}_D[v])_{-\frac{1}{2},\frac{1}{2}}$$

    $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$: duality pairing between $H^{-\frac{1}{2}}(\partial D)$ and $H^{\frac{1}{2}}(\partial D)$.
  - In two dimensions: $\exists ! \widetilde{\varphi}_0$ s.t. $\mathcal{S}_D[\widetilde{\varphi}_0] =$ constant on ∂D and $(\widetilde{\varphi_0}, 1)_{-\frac{1}{2},\frac{1}{2}} = 1$. $\mathcal{S}_D \to \widetilde{\mathcal{S}}_D$:

$$\widetilde{\mathcal{S}}_D[\varphi] = \begin{cases} \mathcal{S}_D[\varphi] & \text{if } (\varphi, 1)_{-\frac{1}{2},\frac{1}{2}} = 0, \\ -1 & \text{if } \varphi = \widetilde{\varphi}_0. \end{cases}$$

Plasmonic resonances for nanoparticles

- Symmetrization technique for Neumann-Poincaré operator $\mathcal{K}_D^*$:
  - Spectrum $\sigma(\mathcal{K}_D^*)$ discrete in $]-1/2, 1/2[$;
  - Ellipse: $\pm\frac{1}{2}(\frac{a-b}{a+b})^j$, elliptic harmonics ($a, b$: long and short axis).
  - Ball: $\frac{1}{2(2j+1)}$, spherical harmonics.
  - Twin property in two dimensions;
  - (λ_j, φ_j), $j = 0, 1, 2, \ldots$: eigenvalue and normalized eigenfunction pair of $\mathcal{K}_D^*$ in $\mathcal{H}^*(\partial D)$; $\lambda_j \in]-\frac{1}{2}, \frac{1}{2}]$ and $\lambda_j \to 0$ as $j \to \infty$;
  - φ_0: eigenfunction associated to $1/2$ ($\widetilde{\varphi}_0$ multiple of φ_0);
  - Spectral decomposition formula in $H^{-1/2}(\partial D)$,

$$\mathcal{K}_D^*[\psi] = \sum_{j=0}^{\infty} \lambda_j (\psi, \varphi_j)_{\mathcal{H}^*} \varphi_j.$$

Plasmonic resonances for nanoparticles

- u^i: incident plane wave; Helmholtz equation:

$$\begin{cases} \nabla\cdot\Big(\varepsilon_m\chi(\mathbb{R}^d \setminus \bar{D}) + \varepsilon_c(\omega)\chi(\overline{D})\Big)\nabla u + \omega^2 u = 0, \\ u^s := u - u^i \text{ satisfies the outgoing radiation condition.} \end{cases}$$

- Uniform small volume expansion[3] with respect to the contrast: $D = z + \delta B$, $\delta \to 0$, $|x - z| \gg 2\pi/k_m$,

$$u^s = -M(\lambda(\omega), D)\nabla_z G_{k_m}(x-z)\cdot\nabla u^i(z) + O\Big(\frac{\delta^{d+1}}{\mathrm{dist}(\lambda(\omega), \sigma(\mathcal{K}_D^*))}\Big).$$

- G_{k_m}: outgoing fundamental solution to $\Delta + k_m^2$; $k_m := \omega/\sqrt{\varepsilon_m}$;
- Polarization tensor:

$$M(\lambda(\omega), D) := \int_{\partial D} x(\lambda(\omega)I - \mathcal{K}_D^*)^{-1}[\nu](x)\, ds(x).$$

[3] with P. Millien, M. Ruiz, H. Zhang, Arch. Ration. Mech. Anal., 2016.

Plasmonic resonances for nanoparticles

- Spectral decomposition: (l, m)-entry

$$M_{l,m}(\lambda(\omega), D) = \sum_{j=1}^{\infty} \frac{(\nu_m, \varphi_j)_{\mathcal{H}^*} (\nu_l, \varphi_j)_{\mathcal{H}^*}}{(1/2 - \lambda_j)(\lambda(\omega) - \lambda_j)}.$$

- $(\nu_m, \varphi_0)_{\mathcal{H}^*} = 0$; φ_0: eigenfunction of $\mathcal{K}_D^*$ associated to 1/2.
- Quasi-static plasmonic resonance: $\mathrm{dist}(\lambda(\omega), \sigma(\mathcal{K}_D^*))$ minimal ($\Re e\, \varepsilon_c(\omega) < 0$).

Plasmonic resonances for nanoparticles

- Scattering amplitude:

$$u^s(x) = -ie^{-\frac{\pi i}{4}} \frac{e^{ik_m|x|}}{\sqrt{8\pi k_m|x|}} A_\infty[D, \varepsilon_c, \varepsilon_m, \omega](\theta, \theta') + o(|x|^{-\frac{1}{2}}),$$

  $|x| \to \infty$; θ, θ': incident and scattered directions.

- Scattering cross-section:

$$Q^s[D, \varepsilon_c, \varepsilon_m, \omega](\theta') := \int_0^{2\pi} \left| A_\infty[D, \varepsilon_c, \varepsilon_m, \omega](\theta, \theta') \right|^2 d\theta.$$

- Enhancement of Q^s at plasmonic resonances:

$$Q^s \propto \left| \mathrm{tr}(M(\lambda(\omega), D)) \right|^2.$$

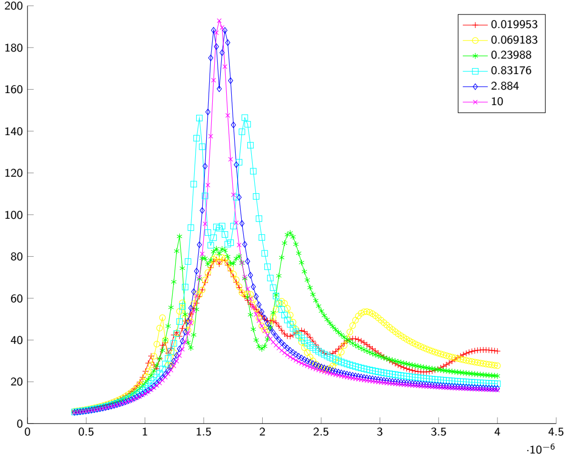
Plasmonic resonances for nanoparticles

- Quasi-plasmonic resonances for multiple particles: D_1 and D_2: $\mathcal{C}^{1,\alpha}$-bounded domains; $\mathrm{dist}(D_1, D_2) > 0$; $\nu^{(1)}$ and $\nu^{(2)}$: outward normal vectors at ∂D_1 and ∂D_2.
- Neumann-Poincaré operator $\mathbb{K}^*_{D_1 \cup D_2}$ associated with $D_1 \cup D_2$:

$$\mathbb{K}^*_{D_1 \cup D_2} := \begin{pmatrix} \mathcal{K}^*_{D_1} & \frac{\partial}{\partial \nu^{(1)}} \mathcal{S}_{D_2} \\ \frac{\partial}{\partial \nu^{(2)}} \mathcal{S}_{D_1} & \mathcal{K}^*_{D_2} \end{pmatrix}.$$

- Symmetrization of $\mathbb{K}^*_{D_1 \cup D_2}$.
- Behavior of the eigenvalues of $\mathbb{K}^*_{D_1 \cup D_2}$ as $\mathrm{dist}(D_1, D_2) \to 0$.

Plasmonic resonances for nanoparticles



Norm of the polarization tensor for two disks for various separating distances.

Plasmonic resonances for nanoparticles

- Two nearly touching disks: infinite number of quasi-static plasmonic resonances.

$$\lambda_j = \pm \frac{1}{2} e^{-2|j|\xi}, \xi = \sinh^{-1}\left(\sqrt{\frac{\delta}{R}\left(1 + \frac{\delta}{4R}\right)}\right);$$

- R: radius of the particles; δ: separating distance.
- Blow-up of the electric field between the particles at plasmonic resonances[4]:

$$\nabla u = O\left(\frac{R}{\delta} \times \frac{1}{\Im m \lambda(\omega)}\right) \qquad \text{in two dimensions.}$$

- Singular interaction between nearly touching plasmonic nanoparticles: applications in nanosensing.

[4] with M. Putinar, M. Ruiz, S. Yu, H. Zhang, J. Math. Pures Appl., 2018.

Plasmonic resonances for nanoparticles

- Fully analytic solution for two plasmonic spheres[5].
- Capture analytically the singularity in the gap between the plasmonic spheres.
- Efficient and accurate hybrid scheme valid for arbitrary number of plasmonic spheres which can be nearly touching.
- Key idea: clarify the connection between Transformation optics and the method of image charges.

[5] with S. Yu, SIAM Rev., 2018.

Plasmonic resonances for nanoparticles

- Uniform incident field $(0, 0, E_0)$ in the direction of the z-axis. In the case of the x or y-axis, a high field concentration in the gap does not happen.
- Method of image charges: infinite series of image charges of strength $\pm u_k$ at $\mathbf{z}_k := (0, 0, \pm z_k)$

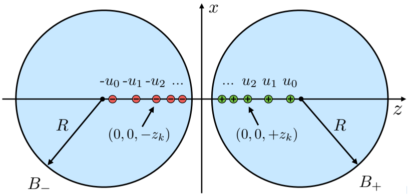
$$u(\mathbf{r}) = \sum_{k=0}^{\infty} u_k (G(\mathbf{r} - \mathbf{z}_k) - G(\mathbf{r} + \mathbf{z}_k));$$

$\tau = (\varepsilon_c - 1)/(\varepsilon_c + 1) = 1/(2\lambda)$, $s = \cosh^{-1}(\delta/R)$ and $\alpha = R \sinh s$.

$$z_k = \alpha \coth(ks + s + t_0), \quad u_k = \tau^k \frac{\sinh(s + t_0)}{\sinh(ks + s + t_0)}.$$

t_0 s.t. $z_0 = \alpha \coth(s + t_0)$.

- Not valid for plasmonic spheres due to non-convergence.



Plasmonic resonances for nanoparticles

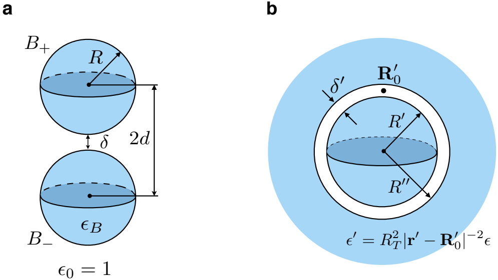
- Transformation Optics (TO) basis:

$$\mathcal{M}_{n,\pm}^m(\mathbf{r}) = |\mathbf{r}' - \mathbf{R}_0'|(r')^{\pm(n+\frac{1}{2})-\frac{1}{2}} Y_n^m(\theta', \phi'),$$

  Y_n^m: spherical harmonics.
- TO solution:

$$u(\mathbf{r}) = -E_0 z + \sum_{n=0}^{\infty} A_n \big(\mathcal{M}_{n,+}^0(\mathbf{r}) - \mathcal{M}_{n,-}^0(\mathbf{r})\big).$$

- TO solution: not fully analytic.



Plasmonic resonances for nanoparticles

- Convert the image charge solution into a Transformation optics solution: for $\mathbf{r} \in \mathbb{R}^3 \setminus (B_+ \cup B_-)$,

$$u_k G(\mathbf{r} \mp \mathbf{z}_k) = \frac{\sinh(s + t_0)}{4\pi\alpha} \sum_{n=0}^{\infty} \left[\tau e^{-(2n+1)s}\right]^k e^{-(2n+1)(s+t_0)} \mathcal{M}_{n,\pm}^0(\mathbf{r}).$$

- If $|\tau| \approx 1$, the following approximation for the electric potential $V(\mathbf{r})$ holds: for $\mathbf{r} \in \mathbb{R}^3 \setminus (B_+ \cup B_-)$,

$$V(\mathbf{r}) \approx -E_0 z + \sum_{n=0}^{\infty} \widetilde{A}_n \Big(\mathcal{M}_{n,+}^0(\mathbf{r}) - \mathcal{M}_{n,-}^0(\mathbf{r})\Big);$$

$\widetilde{A}_n$: explicit.

Plasmonic resonances for nanoparticles

- Approximate resonance condition given by

$$\sum_n (\tau - e^{(2n+1)s})^{-1} = 0.$$

- Eigenvalue estimates:

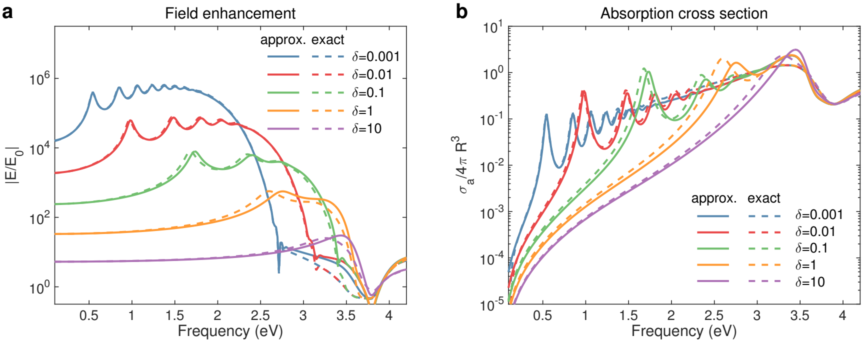
$$\frac{1}{2} e^{-(2n+3)s} < \lambda_n < \frac{1}{2} e^{-(2n+1)s}, \quad s = \sqrt{\frac{\delta}{R}} + O(\delta^{3/2}).$$

- Blow-up of the electric field in the gap at the plasmonic resonances:

$$\nabla u = O\Big(\frac{1}{(\delta/R)^{3/2} \ln(R/\delta)} \times \frac{1}{\Im m \lambda(\omega)}\Big).$$

- Non-local effect (quantum origin) $\Rightarrow$ in the touching case, gap distance effectively non-zero.

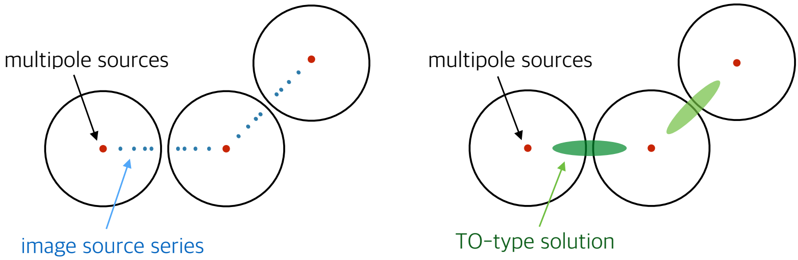
Plasmonic resonances for nanoparticles



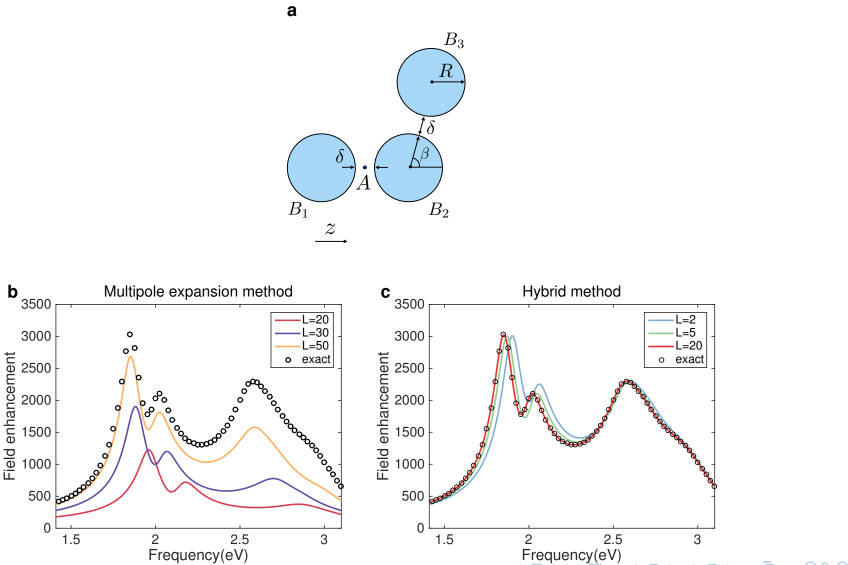
Plasmonic resonances for nanoparticles

- Efficient and accurate hybrid scheme valid for arbitrary number of plasmonic spheres which can be nearly touching.
- Modify Cheng and Greengard's hybrid scheme by replacing image source series with their TO versions.
- 2000 times faster than the multipole expansion method.

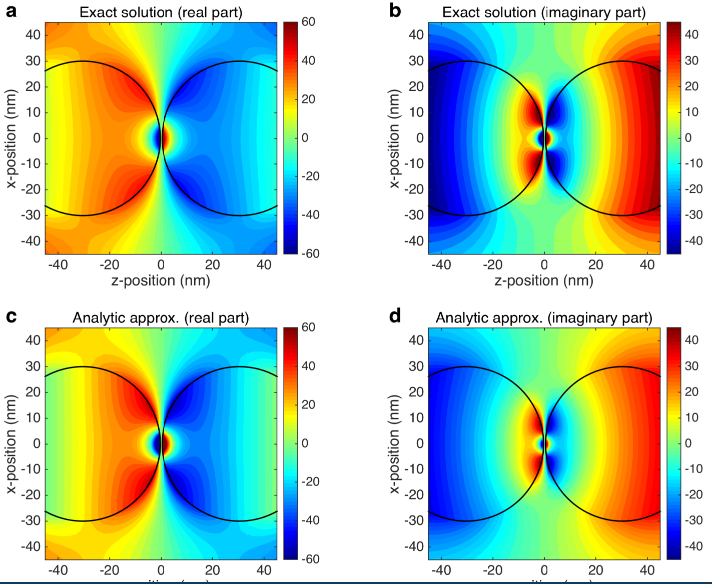
Plasmonic resonances for nanoparticles



Plasmonic resonances for nanoparticles



Plasmonic resonances for nanoparticles

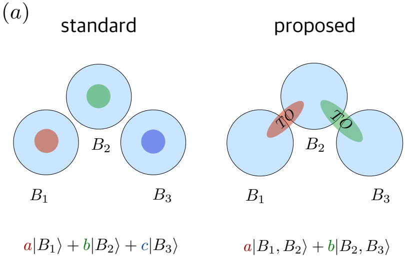


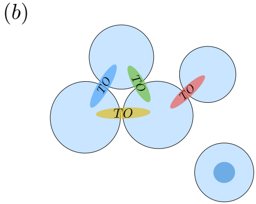
Plasmonic resonances for nanoparticles

- Singular hybridization (SH) model for plasmons of strongly interacting many-particle systems[6].
- SH model combines the advantages of both the hybridization and TO approaches, thus providing a simple and intuitive picture when the particles are close-to-touching.
- SH model leads to new physical insights into the relation between geometry and plasmons: how global and local information of the system's complex geometry are encoded into the spectrum of the plasmons.
- SH model enables us to decompose the spectrum into singularly and regularly shifted parts. The singular (resp. regular) part is controlled by local (resp. global) features of the geometry.
- SH model informs us on how we can control them in a systematic way, opening up new degrees of freedom for light manipulation at the nanoscale.

[6] with S. Yu, submitted, 2018.

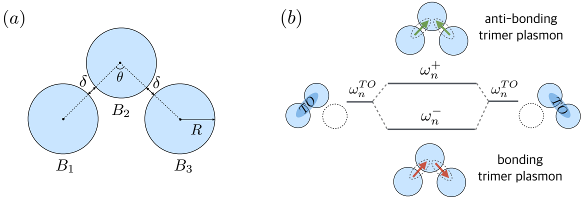
Plasmonic resonances for nanoparticles





A more general case

Plasmonic resonances for nanoparticles



Plasmonic resonances for nanoparticles

- Coupled mode equations for the hybridization of dimer plasmons:

$$\begin{bmatrix} (\omega_n^{TO})^2 & \Delta_n \\ \Delta_n & (\omega_n^{TO})^2 \end{bmatrix} \begin{bmatrix} a_n \\ b_n \end{bmatrix} = \omega^2 \begin{bmatrix} a_n \\ b_n \end{bmatrix}.$$

- Δ_n: coupling between the two TO modes.
- Spectral theory of the Neumann–Poincaré operator $\Rightarrow$ hybrid modes for the trimer:

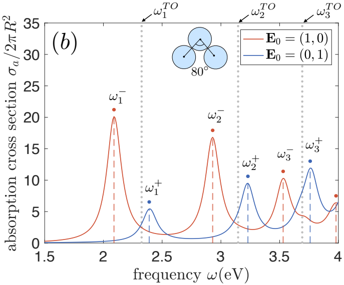
$$|\omega_n^{\pm}\rangle \approx \frac{1}{\sqrt{2}}\Big(|\omega_n^{TO}(B_1, B_2)\rangle \mp |\omega_n^{TO}(B_2, B_3)\rangle\Big), \quad n = 1, 2, 3, \cdots,$$

and their resonance frequencies

$$\omega_n^{\pm} \approx \omega_n^{TO} \pm \Delta_n, \quad n = 1, 2, 3, \cdots.$$

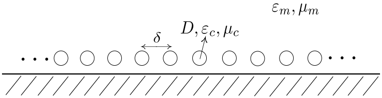
- As the bonding angle between the two gap-plasmons decreases, the coupling strength Δ_n increases, which is to be expected since the two gaps get closer.

Plasmonic resonances for nanoparticles



Plasmonic resonances for nanoparticles

- Metasurfaces:



- Effective boundary condition: $u_{\mathrm{app}} + Z(\omega) \dfrac{\partial u_{\mathrm{app}}}{\partial x_2} = 0.$
- Effective impedance[7]:

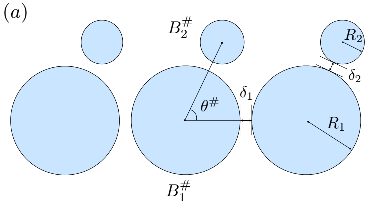
$$Z(\omega) = \delta \sum_{j=1}^{+\infty} \frac{(\varphi_j, \nu_2)^2}{\lambda(\omega) - \lambda_j} \frac{1}{\frac{1}{2} - \lambda_j}.$$

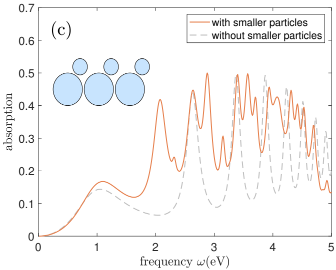
- (φ_j, λ_j): eigenvectors and eigenvalues of the associated Neumann-Poincaré operator.
- Pointwise estimate in the far-field: $u = u_{\mathrm{app}}(1 + O(\delta))$.

[7] with M. Ruiz, S. Yu, W. Wu, H. Zhang, Proc. Royal Soc. A, 2016

Plasmonic resonances for nanoparticles

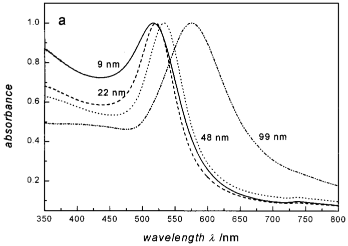
- Broadband metasurfaces: singular hybridization $\Rightarrow$ dense spectrum.





Plasmonic resonances for nanoparticles

- $\mathcal{K}_D^*$: scale invariant $\Rightarrow$ Quasi-static plasmonic resonances: size independent.
- Analytic formula for the first-order correction to quasi-static plasmonic resonances in terms of the particle's characteristic size δ:



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Plasmonic resonances for nanoparticles

- Helmholtz equation:

$$\begin{cases} \nabla \cdot \Big(\varepsilon_m \chi(\mathbb{R}^d \setminus \bar{D}) + \varepsilon_c(\omega)\chi(\overline{D}) \Big) \nabla u + \omega^2 u = 0, \\ u^s := u - u^i \text{ satisfies the outgoing radiation condition.} \end{cases}$$

u^i: incident plane wave; $k_m := \omega\sqrt{\varepsilon_m}$, $k_c := \omega\sqrt{\varepsilon_c(\omega)}$.

- Integral formulation on ∂D:

$$\begin{cases} \mathcal{S}_D^{k_c}[\phi] - \mathcal{S}_D^{k_m}[\psi] = u^i, \\ \varepsilon_c\big(\frac{I}{2} - (\mathcal{K}_D^{k_c})^*\big)[\phi] - \varepsilon_m\big(\frac{I}{2} + (\mathcal{K}_D^{k_m})^*\big)[\psi] = \varepsilon_m \partial u^i/\partial \nu. \end{cases}$$

- Operator-valued function $\delta \mapsto \mathcal{A}_\delta(\omega) \in \mathcal{L}(\mathcal{H}^*(\partial B), \mathcal{H}^*(\partial B))$:

$$\mathcal{A}_\delta(\omega) = \overbrace{(\lambda(\omega)I - \mathcal{K}_B^*)}^{\mathcal{A}_0(\omega)} + (\omega\delta)^2 \mathcal{A}_1(\omega) + O((\omega\delta)^3).$$

- Quasi-static limit:

$$\mathcal{A}_0(\omega)[\psi] = \sum_{j=0}^{\infty} \tau_j(\omega)(\psi, \varphi_j)_{\mathcal{H}^*}\varphi_j, \quad \tau_j(\omega) := \frac{1}{2}(\varepsilon_m + \varepsilon_c(\omega)) - (\varepsilon_c(\omega) - \varepsilon_m)\lambda_j.$$

Plasmonic resonances for nanoparticles

- Shift in the plasmonic resonances[8]:

$$\arg\min_{\omega} \Big| \frac{1}{2}\big(\varepsilon_m + \varepsilon_c(\omega)\big) - \big(\varepsilon_c(\omega) - \varepsilon_m\big)\lambda_j + (\omega\delta)^2 \tau_{j,1} \Big|$$

- $\tau_{j,1} := (\mathcal{A}_1(\omega)[\varphi_j], \varphi_j)_{\mathcal{H}^*}$.

[8] with P. Millien, M. Ruiz, H. Zhang, Arch. Ration. Mech. Anal., 2016.

Minnaert resonances for bubbles

- Oscillation: Spherical gas bubbles in liquid oscillate at a natural frequency called the Minnaert resonance.
- Sub-wavelength resonance: Associated wavelength several orders of magnitude larger than bubble size.
- Scattering: Bubbles are very strong monopole scatterers of sound.

Minnaert resonances for bubbles

A single bubble D in a liquid:

- Model:

$$\begin{cases} \nabla \cdot \dfrac{1}{\rho} \nabla u + \dfrac{\omega^2}{\kappa} u = 0 \quad \text{in} \quad \mathbb{R}^d \setminus \overline{D}, \\ \nabla \cdot \dfrac{1}{\rho_b} \nabla u + \dfrac{\omega^2}{\kappa_b} u = 0 \quad \text{in} \quad D, \\ u|_+ = u|_- \quad \text{on} \quad \partial D, \\ \dfrac{1}{\rho} \dfrac{\partial u}{\partial \nu}\Big|_+ = \dfrac{1}{\rho_b} \dfrac{\partial u}{\partial \nu}\Big|_- \quad \text{on} \quad \partial D, \\ u^s := u - u^{in} \text{ satisfies the Sommerfeld radiation condition.} \end{cases}$$

- ρ_b, ρ: densities inside and outside the bubble; κ_b, κ: bulk moduli inside and outside the bubble.
- High contrast: $\delta := \rho_b/\rho \ll 1$; $\tau := \kappa_b/\kappa = O(1)$.
- $v = \sqrt{\kappa/\rho}$; $v_b = \sqrt{\kappa_b/\rho_b}$; $k = \omega\sqrt{\rho/\kappa}$.

Minnaert resonances for bubbles

- Minnaert resonance frequency for a bubble of arbitrary shape[9]:

$$\omega_M(\delta) = \left(\sqrt{\frac{\mathrm{Cap}_D}{|D|}} v_b \sqrt{\delta} - i \frac{\mathrm{Cap}_D^2 v_b^2}{8\pi v |D|} \delta + O(\delta^{\frac{3}{2}}) \right).$$

- Capacity $\mathrm{Cap}_D := \int_{\partial D} \mathcal{S}_D^{-1}[1] \, d\sigma.$
- Monopole approximation near the Minnaert resonance frequency:

$$u^s(x) = g(\omega, \delta, D)(1 + O(\omega) + O(\delta) + o(1)) u^{in}(x_0) G_k(x, x_0).$$

- Scattering coefficient g:

$$g(\omega, \delta, D) = \frac{\mathrm{Cap}_D}{1 - (\frac{\omega_M}{\omega})^2 + i\gamma}.$$

- Scattering enhancement near the Minnaert resonance frequency.

[9] with B. Fitzpatrick, D. Gontier, H. Lee, H. Zhang, Ann. IHP C, 2018.

Minnaert resonances for bubbles

- Integral formulation: $\mathcal{A}(\omega,\delta)[\Psi] = F$;
-

$$\mathcal{A}(\omega,\delta) = \begin{pmatrix} \mathcal{S}_D^{k_b} & -\mathcal{S}_D^{k} \\ -\frac{1}{2} + \mathcal{K}_D^{k_b,*} & -\delta(\frac{1}{2} + (\mathcal{K}_D^{k})^*) \end{pmatrix}, \ \Psi = \begin{pmatrix} \psi_b \\ \psi \end{pmatrix}, \ F = \begin{pmatrix} u^{in} \\ \delta \frac{\partial u^{in}}{\partial \nu} \end{pmatrix}.$$

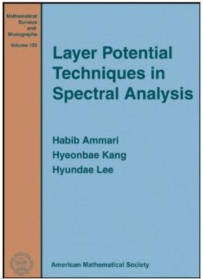
- 0: characteristic value of the limiting operator-valued function:

$$\mathcal{A}_0(0,0) = \begin{pmatrix} \mathcal{S}_D & -\mathcal{S}_D \\ -\frac{1}{2} + \mathcal{K}_D^* & 0 \end{pmatrix}.$$

- Gohberg-Sigal theory:
  - Generalization of argument principle.
  - V: complex neighborhood of 0:

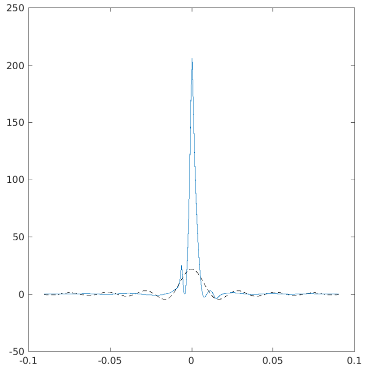
$$\omega_M(\delta) = \frac{1}{2\pi i}\mathrm{tr}\int_{\partial V} \omega \mathcal{A}(\omega,\delta)^{-1} \frac{\partial}{\partial \omega}\mathcal{A}(\omega,\delta)\, d\omega.$$

  - Muller's method: compute characteristic eigenvalues.



Super-focusing in bubbly media

- Dilute regime: When excited slightly below the Minnaert resonance frequency ω_M a large number of small bubbles acts as an effective medium with high refractive index in which super-focusing and super-resolution is achievable[10]:



[10] with B. Fitzpatrick, D. Gontier, H. Lee, H. Zhang, Proc. Royal Soc. A, 2017.

Super-focusing in bubbly media

- Effective operator[11]:

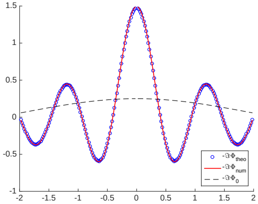
$$\Delta + k^2 + V(x); \qquad V(x) = \frac{1}{(\frac{\omega_M}{\omega})^2 - 1} \Lambda \widetilde{V}(x).$$

  - Λ: depends only on the size and number of the bubbles;
  - $\widetilde{V}$: depends only on the distribution of the centers of the bubbles.
- ω slightly below ω_M: high-contrast refractive index;
- ω slightly above ω_M: negative bulk modulus;
- Effective medium theory: does not hold at $\omega = \omega_M$.

[11] with H. Zhang, SIAM J. Math. Anal., 2017.

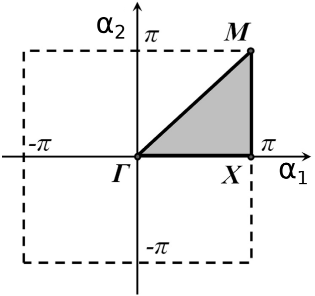
Super-focusing in bubbly media

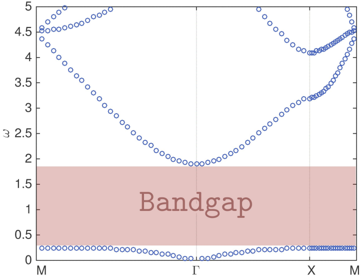
- Mechanism of super-focusing in high-contrast media:
  - Mixing of resonant modes: intrinsic nature of non-hermitian systems.
  - Sub-wavelength resonance modes excited $\Rightarrow$ dominate over the other ones in the expansion of the Green function.
  - Imaginary part of the Green function may have sharper peak than the one of G due to the excited sub-wavelength resonant modes.
  - Sub-wavelength modes: determine the super-resolution.



Sub-wavelength bandgap opening

- Sub-wavelength phononic bandgaps[12]: Due to the phenomena of sub-wavelength resonance, bubbles can be used to create phononic crystals in which low frequency wave propagation is prohibited.
- Sub-wavelength bandgaps: appear slightly above the Minnaert resonance ω_*.
- Super-focusing: appear slightly below the Minnaert resonance ω_*.





[12] with B. Fitzpatrick, H. Lee, S. Yu, H. Zhang, J. Diff. Equat., 2017.

Sub-wavelength bandgap opening

- For $\alpha \in [0, 2\pi)^3$,

$$\mathcal{A}(\omega, \delta, \alpha) = \begin{pmatrix} \mathcal{S}_D^{k_b} & -\mathcal{S}_D^{\alpha,k} \\ -\frac{1}{2} + \mathcal{K}_D^{k_b,*} & -\delta(\frac{1}{2} + (\mathcal{K}_D^{-\alpha,k})^*) \end{pmatrix}, \ \Psi = \begin{pmatrix} \psi_b \\ \psi \end{pmatrix}.$$

- $\mathcal{S}_D^{\alpha,k}$ and $\mathcal{K}_D^{-\alpha,k}$ associated with quasi-periodic Green's function:

$$G^{\alpha,k}(x,y) = \sum_{n \in \mathbb{Z}^3} \frac{e^{i(2\pi n + \alpha)\cdot(x-y)}}{k^2 - |2\pi n + \alpha|^2}.$$

- Characteristic values of $\mathcal{A}(\omega, \delta, \alpha)$:

$$0 \leq \omega_1^\alpha \leq \omega_2^\alpha \leq \dots$$

Sub-wavelength bandgap opening

- ω_0: characteristic value of $\mathcal{A}(\omega, 0)$ iff $(\omega_0/v_b)^2$: Neumann eigenvalue of D or $(\omega_0/v)^2$: Dirichlet eigenvalue of $Y \setminus D$ with α-quasiperiodicity on ∂Y.
- For any δ sufficiently small, there exists one and only one characteristic value $\omega_0 = \omega_0(\delta)$ in a complex neighborhood of 0 to $\mathcal{A}(\omega, \delta)$.
- $\omega_0(0) = 0$ and $\omega_0(\delta)$ depends on δ continuously.
- Asymptotic behavior of ω_1^α:
  - For $\alpha \neq 0$ and sufficiently small δ,

$$\omega_1^\alpha = \omega_M \sqrt{c_2} + O(\delta^{3/2});$$

  - ω_M: free space Minnaert resonant frequency;
  - $c_2 := \mathrm{Cap}_{D,\alpha}/\mathrm{Cap}_D$;
  - Quasi-periodic capacity:

$$\mathrm{Cap}_{D,\alpha} := -\int_{\partial D} (\mathcal{S}_D^{\alpha,0})^{-1}[1]\, d\sigma.$$

Sub-wavelength bandgap opening

- Sub-wavelength bandgap opening:
  - For every sufficiently small $\epsilon > 0$, there exists $\delta_0 > 0$ and $\tilde{\omega} > \omega_1^* + \epsilon$ s.t., for $\delta < \delta_0$,

$$[\omega_1^* + \epsilon, \tilde{\omega}] \subset [\max_\alpha \omega_1^\alpha, \min_\alpha \omega_2^\alpha].$$

- D: symmetric with respect to planes $\{(x_1, x_2, x_3) : x_j = 0\}$, $j = 1, 2, 3$.
- $\mathrm{Cap}_{D,\alpha}$ and ω_1^α attain their maxima at $\alpha^* = (\pi, \pi, \pi)$ (ω_1^α attained at the corner M of the Brillouin zone).
- $v = v_b$: wave speed inside the bubble is equal to the one outside.
- For $\epsilon > 0$ small enough,

$$\mathrm{Cap}_{D,\alpha^*+\epsilon\tilde{\alpha}} = \mathrm{Cap}_{D,\alpha^*} + \epsilon^2 \Lambda_D^{\tilde{\alpha}} + O(\epsilon^4).$$

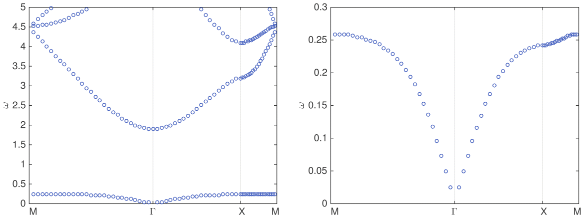
- $\Lambda_D^{\tilde{\alpha}}$: negative semi-definite quadratic function of $\tilde{\alpha} \Rightarrow$

$$\frac{v_b^2}{|D|} \Lambda_D^{\tilde{\alpha}} = - \sum_{1 \le i,j \le 3} \lambda_{ij} \tilde{\alpha}_i \tilde{\alpha}_j.$$

- (λ_{ij}): symmetric and positive semi-definite.

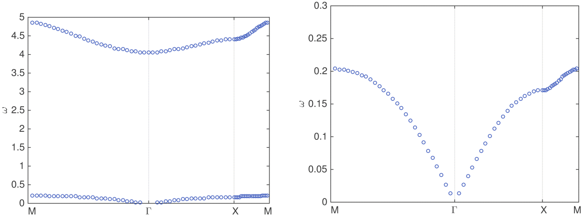
Sub-wavelength bandgap opening

- Dilute regime:
  - $\omega_* \approx \omega_M$.
  - Effective medium theory near Minnaert resonant frequency ω_*.
  - High contrast effective medium $\Rightarrow$ super-focusing.
  - Negative effective medium $\Rightarrow$ sub-wavelength bandgap opening.
- Band structure of a square array of circular bubbles with radius $R = 0.05$ and contrast $\delta^{-1} = 5000$:



Sub-wavelength bandgap opening

- Non-dilute regime:
  - High-frequency homogenization.
- Band structure of a square array of circular bubbles with radius $R = 0.25$ and contrast $\delta^{-1} = 1000$:



Sub-wavelength bandgap opening

- s: period of the crystal; $\delta = O(s^2)$.
- $\omega_*^s = (1/s)\omega_*^1$; Critical frequencies $= O(1)$ as $s \to 0$.
- Near the critical frequency ω_*^s: eigenfunctions can be decomposed into two parts[13]:
  - One part: slowly varying and satisfies a homogenized equation;
  - Second part: periodic across each elementary crystal cell and is varying.
- $(\omega_*^s)^2 - \omega^2 = O(s^2)$; Asymptotic of Bloch eigenfunction $u_{1,s}^{\alpha^*/s+\tilde{\alpha}}$:

$$u_{1,s}^{\alpha^*/s+\tilde{\alpha}}(x) = e^{i\tilde{\alpha}\cdot x} S\left(\frac{x}{s}\right) + O(s);$$

- Macroscopic plane wave $e^{i\tilde{\alpha}\cdot x}$ satisfies:

$$\sum_{1\leq i,j\leq 3} \lambda_{ij}\partial_i\partial_j \hat{u}(x) + \frac{\omega_*^2 - \omega^2}{\delta}\hat{u}(x) = 0.$$

[13] with H. Lee, H. Zhang, submitted, 2018.

Sub-wavelength bandgap opening

- $(\omega_*^s)^2 - \omega^2 = \beta\delta$;
- $\sum_{1\leq i,j\leq 3} \lambda_{ij}\tilde{\alpha}_i\tilde{\alpha}_j = \beta + O(s^2)$:
  - $\beta > 0 \Rightarrow$ plane wave Bloch eigenvalues:
    - Homogenized equation for the bubbly phononic crystal;
    - Microscopic field: periodic and varies on the scale of s;
    - Microscopic oscillations of the field at the period of the crystal justify the super-focusing phenomenon.
  - $\beta < 0 \Rightarrow$ exponentially growing or decaying functions $\Rightarrow$ bandgap opening.

Sub-wavelength cavities

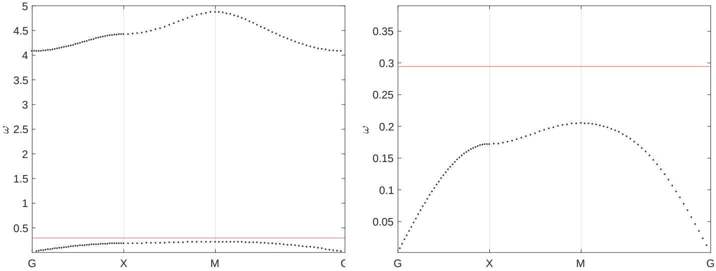
- Sub-wavelength localized modes $\Leftarrow$ increase the radius of one bubble (from R to $R+\epsilon$) to create a detuned resonator with an upward shifted resonance frequency (within the sub-wavelength bandgap).
- As $\epsilon \to 0$[14],

$$\omega^{\epsilon} - \omega_* \approx \exp\Big(- \frac{c_\delta}{2\epsilon} \frac{\frac{R^2}{2}\omega_*(-\ln \omega^*)}{\big(\frac{R^2}{2}(\omega_*)^2(-\ln \omega_*) + \delta\big)}\Big);$$

c_δ: positive constant.

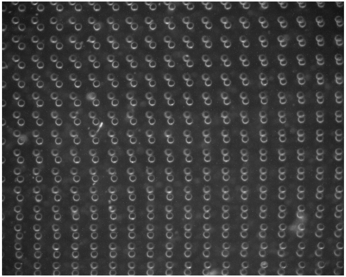
[14] with B. Fitzpatrick, E. Orvehed Hiltunen, S. Yu, submitted, 2018.

Sub-wavelength cavities



Double-negative acoustic metamaterials

- Bubble dimers ⇒ double-negative acoustic metamaterials[15]:



[15] with B. Fitzpatrick, H. Lee, S. Yu, H. Zhang, submitted, 2018.

Double-negative acoustic metamaterials

- Capacitance coefficients:
  - $D = D_1 \cup D_2$; ψ_1, $\psi_2 \in L^2(\partial D)$:

$$\mathcal{S}_D^0[\psi_1] = \begin{cases} 1 & \text{on } \partial D_1, \\ 0 & \text{on } \partial D_2, \end{cases} \qquad \mathcal{S}_D^0[\psi_2] = \begin{cases} 0 & \text{on } \partial D_1, \\ 1 & \text{on } \partial D_2. \end{cases}$$

  - $\ker\left(-\frac{1}{2}I + \mathcal{K}_D^{0,*}\right) = \text{span}\ \{\psi_1, \psi_2\}$.
  - $\psi_1 \pm \psi_2$: symmetric and anti-symmetric modes.
  - Capacitance coefficients matrix $C = (C_{ij})$:

$$C_{ij} := -\int_{\partial D_j} \psi_i, \quad i, j = 1, 2.$$

  - C: positive definite and symmetric.
  - D_1 and D_2 identical balls: $C_{11} = C_{22}$, $C_{12} = C_{21}$, $C_{11} > 0$, and $C_{12} < 0$.
  - Explicit formulas: bispherical coordinates.

Double-negative acoustic metamaterials

- Resonances for a dimer consisting of two identical bubbles:
  - Two quasi-static resonances with positive real part for the bubble dimer D.
  - As $\delta \to 0$,

$$\omega_{M,1} = \sqrt{(C_{11} + C_{12})} v_b \sqrt{\delta} - i\tau_1 \delta + O(\delta^{3/2}),$$
$$\omega_{M,2} = \sqrt{(C_{11} - C_{12})} v_b \sqrt{\delta} + \delta^{3/2} \hat{\eta}_1 + i\delta^2 \hat{\eta}_2 + O(\delta^{5/2}).$$

  - $\hat{\eta}_1$ and $\hat{\eta}_2$: real numbers determined by D, v, and v_b;

$$\tau_1 = \frac{v_b^2}{4\pi v}(C_{11} + C_{12})^2.$$

  - Resonances $\omega_{M,1}$ and $\omega_{M,2}$: hybridized resonances of the bubble dimmer D.

Double-negative acoustic metamaterials

- Bubble dimer: approximated as a point scatterer with monopole and dipole modes.
- For $\omega = O(\delta^{1/2})$ and $\delta \to 0$, $|x|$: sufficiently large,

$$
\begin{aligned}
u(x) - u^{in}(x) &= g^0(\omega) u^{in}(0) G_k(x,0) \\
&\quad + \nabla u^{in}(0) \cdot g^1(\omega) \nabla G_k(x,0) + O(\delta |x|^{-1}).
\end{aligned}
$$

- Scattering coefficients:

$$
g^0(\omega) = \frac{C(1,1)}{1 - \omega_{M,1}^2/\omega^2}(1 + O(\delta^{1/2})), \quad C(1,1) := C_{11} + C_{12} + C_{21} + C_{22};
$$

$$
g^1(\omega) = (g^1_{ij}(\omega));
$$

$$
g^1_{ij}(\omega) = \int_{\partial D} (\mathcal{S}_D^0)^{-1}[x_i](y) y_j - \frac{\delta v_b^2}{\omega^2 |D| (1 - \omega_{M,2}^2/\omega^2)} P^2 \delta_{i,1}\delta_{j,1};
$$

$$
P := \int_{\partial D} y_1 (\psi_1 - \psi_2) d\sigma(y).
$$

Double-negative acoustic metamaterials

- Effective medium theory:
  - Bubble dimers: $\{z_i^N : 1 \leq i \leq N\}$; orientation $\{d_i^N : 1 \leq i \leq N\}$.
  - Scattering of an incident acoustic plane wave u^{in} by N identical bubble dimers with random orientations:

$$
\begin{cases}
\nabla \cdot \frac{1}{\rho} \nabla u^N + \frac{\omega^2}{\kappa} u^N = 0 \quad \text{in } \mathbb{R}^3 \backslash D^N, \\
\nabla \cdot \frac{1}{\rho_b} \nabla u^N + \frac{\omega^2}{\kappa_b} u^N = 0 \quad \text{in } D^N, \\
u_+^N - u_-^N = 0 \quad \text{on } \partial D^N, \\
\frac{1}{\rho} \frac{\partial u^N}{\partial \nu} \bigg|_+ - \frac{1}{\rho_b} \frac{\partial u^N}{\partial \nu} \bigg|_- = 0 \quad \text{on } \partial D^N, \\
u^N - u^{in} : \ \text{Sommerfeld radiation condition.}
\end{cases}
$$

  - System of boundary integral equations: $\mathcal{A}^N(\omega, \delta)[\Psi^N] = F^N$.

Double-negative acoustic metamaterials

- Assumptions:
  - s: characteristic size of a bubble dimer; $sN = \Lambda$ for some positive number $\Lambda > 0$.
  - Volume fraction of the bubble dimers is of the order of $s^3 N$.
  - Bubble dimers: very dilute with the average distance between neighboring dimers being of the order of $N^{-1/3}$.

Double-negative acoustic metamaterials

- $u^N(x) \to u(x)$ uniformly for $x \in \Omega_N$.
- u: homogenized model

$$\nabla \cdot \left(I - \Lambda \tilde{g}^1 \tilde{B} \right) \nabla u(x) + (k^2 - \Lambda \tilde{g}^0 \tilde{V}) u = 0, \quad \text{in } \Omega.$$

- $\tilde{B} \in C^1(\bar{\Omega})$: s.t. for $f \in (C^{0,\alpha}(\Omega))^3$ with $0 < \alpha \leq 1$,

$$\max_{1 \leq j \leq N} \Big| \max_{1 \leq j \leq N} \Big| \frac{1}{N} \sum_{i \neq j} (f(z_i^N) \cdot d_i^N)(d_i^N \cdot \nabla G_k(z_i^N, z_j^N))$$

$$- \int_\Omega f(y) \tilde{B} \nabla_y G_k(y, z_j^N) dy \Big| \leq C \frac{1}{N^{\frac{\alpha}{3}}} \|f\|_{C^{0,\alpha}(\Omega)};$$

- $\tilde{V} \in C^1(\bar{\Omega})$: s.t. for any $f \in C^{0,\alpha}(\Omega)$ with $0 < \alpha \leq 1$,

$$\max_{1 \leq j \leq N} \Big| \frac{1}{N} \sum_{i \neq j} G_k(z_j^N, z_i^N) f(z_i^N) - \int_\Omega G_k(z_j^N, y) \tilde{V}(y) f(y) dy \Big|$$

$$\leq C \frac{1}{N^{\frac{\alpha}{3}}} \|f\|_{C^{0,\alpha}(\Omega)}$$

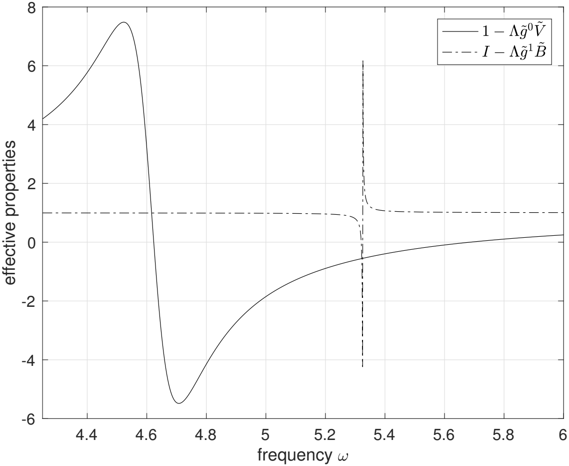
for some constant C independent of N.

Double-negative acoustic metamaterials

- $\delta = \mu^2 s^2$;
- $$\omega_{M,1} = v_b \mu \sqrt{(C_{11} + C_{12})}, \quad \omega_{M,2} = v_b \mu \sqrt{(C_{11} - C_{12})};$$
- Monopole and dipole coefficients:
$$\tilde{g}^0 = \frac{2(C_{11} + C_{12})}{1 - \omega_{M,1}^2/\omega_{M,2}^2}, \quad \tilde{g}^1 = \frac{\mu^2 v_b^2}{2|D|\omega_{M,2}(\mu^3 \hat{\eta}_1 - a)} P^2.$$
- Bubble dimers distributed s.t. $\tilde{B}$: positive matrix with $\tilde{B}(x) \geq C > 0$ for some constant C for all $x \in \Omega \Rightarrow$ both the matrix $I - \Lambda \tilde{g}^1 \tilde{B}$ and the scalar function $k^2 - \Lambda \tilde{g}^0 \tilde{V}$: negative.
- Effective double-negative medium with both negative mass density and negative bulk modulus.

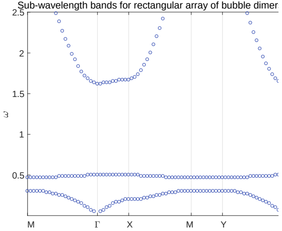
Double-negative acoustic metamaterials

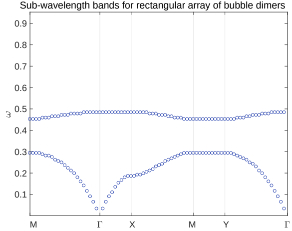
- Effective properties:



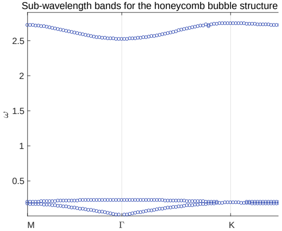
Honeycomb-lattice Minnaert bubbles

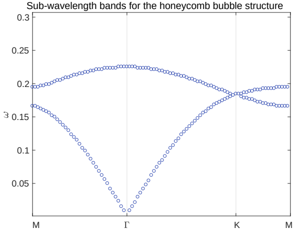
- Rectangular array of bubble dimers:





- Honeycomb-lattice:





Honeycomb-lattice Minnaert bubbles

- At $\alpha = \alpha^*$, the first Bloch eigenfrequency $\omega^* := \omega(\alpha^*)$ of multiplicity 2.
- Conical behavior of sub-wavelength bands[16]: The first band and the second band form a Dirac cone at α^*, i.e.,

$$\omega_1(\alpha) = \omega(\alpha^*) - \lambda|\alpha - \alpha^*|[1 + O(|\alpha - \alpha^*|)],$$
$$\omega_2(\alpha) = \omega(\alpha^*) + \lambda|\alpha - \alpha^*|[1 + O(|\alpha - \alpha^*|)];$$

  $\lambda \neq 0$ for sufficiently small δ.

- Dirac point at $\alpha = \alpha^*$.

[16] with B. Fitzpatrick, H. Lee, E. Orvehed Hiltunen, S. Yu, submitted, 2018.

Concluding remarks

- Sub-wavelength resonances:
  - Helmoholtz resonators.
  - Plasmonic nanoparticles.
  - Minnaert bubbles.
- Effective medium theory:
  - High contrast material $\Rightarrow$ super-focusing.
  - Sub-wavelength bandgap opening $\Rightarrow$ negative materials.
  - Dimers $\Rightarrow$ double-negative materials.
- High-frequency homogenization:
  - Below the critical frequency: super-focusing.
  - Below the critical frequency: Sub-wavelength bandgap opening $\Rightarrow$ negative materials.
- Sub-wavelength cavities; topological properties at sub-wavelength scales.
- Optimal design methodologies.