Supplementary materials: Solitary Waves in Chains of High-index Dielectric Nanoparticles

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Coupled-dipole model for chains of silicon nanoparticles

In the linear case electromagnetic reponse of the dielectric nanoparticle chain can be described by coupled electric dipole (ED) and magnetic dipole (MD) model[1–4], which formulates in the frequency domain as follows:

$$\begin{cases} \mathbf{p}_n = \hat{\alpha}_e(\omega)(\mathbf{E}_n^{(\mathrm{loc})} + \mathbf{E}_n^{(\mathrm{ext})}), \\ \mathbf{m}_n = \hat{\alpha}_m(\omega)(\mathbf{H}_n^{(\mathrm{loc})} + \mathbf{H}_n^{(\mathrm{ext})}), \end{cases} \tag{1}$$

where $\mathbf{m}_n$ and $\mathbf{p}_n$ are magnetic and electric moments induced in the nth particle ($\propto -i\omega t$), $\hat{\alpha}_m$ and $\hat{\alpha}_e$ are magnetic and electric polarizability tensors, electric $\mathbf{E}_n^{(\mathrm{ext})}$ and magnetic $\mathbf{H}_n^{(\mathrm{ext})}$ fields at the position of the nth dipole are produced by external source; local electric $\mathbf{E}_n^{(\mathrm{loc})}$

and magnetic $\mathbf{H}_n^{(\mathrm{loc})}$ fields are produced by all other dipoles in the chain:

$$
\begin{aligned}
\mathbf{E}_n^{(\mathrm{loc})} &= \sum_{j\neq n}\left(\widehat{C}_{nj}\mathbf{p}_j - \widehat{G}_{nj}\mathbf{m}_j\right), \\
\mathbf{H}_n^{(\mathrm{loc})} &= \sum_{j\neq n}\left(\widehat{C}_{nj}\mathbf{m}_j + \widehat{G}_{nj}\mathbf{p}_j\right),
\end{aligned}
\tag{2}
$$

where $\widehat{C}_{nj} = A_{nj}\widehat{I} + B_{nj}(\widehat{\mathbf{r}}_{nj}\otimes\widehat{\mathbf{r}}_{nj})$, $\widehat{G}_{nj} = -D_{nj}\widehat{\mathbf{r}}_{nj}\times\widehat{I}$, $\otimes$ is a dyadic product, $\widehat{I}$ is the unit 3×3 tensor, $\widehat{\mathbf{r}}_{nj}$ is the unit vector in the direction from nth to jth dipole, and

$$
\begin{aligned}
A_{nj} &= e^{ik_hR_{nj}}\left(\frac{k_h^2}{R_{nj}} - \frac{1}{R_{nj}^3} + \frac{ik_h}{R_{nj}^2}\right), \\
B_{nj} &= e^{ik_hR_{nj}}\left(-\frac{k_h^2}{R_{nj}} + \frac{3}{R_{nj}^3} - \frac{3ik_h}{R_{nj}^2}\right), \\
D_{nj} &= e^{ik_hR_{nj}}\left(\frac{k_h^2}{R_{nj}} + \frac{ik_h}{R_{nj}^2}\right),
\end{aligned}
\tag{3}
$$

where $R_{nj} = a|n-j|$ is the distance between the nth and jth dipoles, a is the period of the chain, ε_h is the permittivity of the host medium (in our calculations we take $\varepsilon_h = 1$), and $k_h = \sqrt{\varepsilon_h}\omega/c$ is the host wavenumber. Analytical closed-form dispersion equations for the infinite chains can be obtained by replacing infinite sums in (2) with analytical functions[3,4].

To describe the response of nonspherical nanoparticles analytically (for a certain polarization) we employ approximate expressions for the magnetic and electric polarizabilities of the particles:

$$
\begin{aligned}
\frac{1}{\alpha_m} &= \frac{(\omega_m-\omega)}{\Gamma_m} - i\frac{2}{3}\left(\frac{n_h\omega}{c}\right)^3, \\
\frac{1}{\alpha_e} &= \frac{(\omega_e-\omega)}{\Gamma_e} - i\frac{2}{3}n_h\left(\frac{\omega}{c}\right)^3,
\end{aligned}
\tag{4}
$$

where $\omega_{m,e}$ are MD and ED resonance frequencies, respectively, ω is the external radiation frequency, n_h is the refractive index of the host medium, c is the speed of light, $\Gamma_{m,e}$ and the MD and ED resonance strengths. Exact dependencies $\alpha(\omega)$ can be derived for arbitrary-shaped particles through the multipole decomposition of the field scattered by a single particle[5]. However this approach substantially complicates the derivation of evolution equations (see next section). While fitting $\alpha(\omega)$ with formulae (4) (assuming constant imaginary part at the center frequency of the pulse) provides us with quite simple and accurate approximation.

Evolutions equations

In order to describe the evolution of the induced magnetic and electric moments in time and take into account the resonance frequencies shifts due to the nonlinear response we transform equations (1) into the time domain. After the appropriate normalization we write down the equations for the evolution of the slowly varying transverse components of magnetic $\widetilde{m}$ and electric $\widetilde{p}$ dipole moments of N dielectric nanoparticles (subsript $n = 1..N$ indicates the number of the particle), driven by monochromatic external field in the following form[6]:

$$\begin{cases} -i\dfrac{d\widetilde{m}_n}{d\tau} + (\Omega_m - \widetilde{M}_n - i\gamma_m)\widetilde{m}_n - \beta_m \sum_{j\neq n}(A_{nj}\widetilde{m}_j + \mathrm{sgn}(j-n)D_{nj}\widetilde{p}_j) - \widetilde{H}_n^{(ext)} = 0, \\ -i\dfrac{d\widetilde{p}_n}{d\tau} + (\Omega_e - \widetilde{P}_n - i\gamma_e)\widetilde{p}_n - \beta_e \sum_{j\neq n}(A_{nj}\widetilde{p}_j + \mathrm{sgn}(j-n)D_{nj}\widetilde{m}_j) - \widetilde{E}_n^{(ext)} = 0, \end{cases} \tag{5}$$

where $\tau = \omega_p t$ is the dimensionless time, ω_p is the center frequency of the propagating pulse, $\widetilde{m}_n = \sqrt{\chi_m} m_n$, $\widetilde{p}_n = \sqrt{\chi_e} p_n$, $\widetilde{M}_n = \dfrac{\chi_m}{\omega}|m_n|^2$, $\widetilde{P}_n = \dfrac{\chi_e}{\omega}|p_n|^2$, $\chi_{m,e}$ is the nonlinear coefficient that takes into account the enhancement of electric field intensity inside the particle at MD and ED resonances, $\Omega_{m,e} = \dfrac{\omega_{m,e} - \omega_p}{\omega_p} = \dfrac{k_{m,e} - k_p}{k_p}$ is the relative frequency shift, $k_p = \omega_p/c$, c is the speed of light, $\beta_{m,e} = \dfrac{\Gamma_{m,e}}{\omega_p}$, $\gamma_{m,e} = \beta_{m,e}\dfrac{2}{3}k_p^3$, $\widetilde{H}_n^{(ext)} = \sqrt{\chi}\beta_m H_n^{(ext)}$ and $\widetilde{E}_n^{(ext)} = \sqrt{\chi}\beta_e E_n^{(ext)}$ are normalized electric and magnetic field amplitudes of the external radiation, respectively, at the position of the nth dipole; dipole-dipole interaction constants A_{nj} and D_{nj} are given by formulae (3).

Resonance frequency tuning with nonlinearity

The value of refractive index inside nanoparticle is given by $n = n_L + 2\bar{n}_2|E^{in}|^2$, where n_L is linear refractive index, $\bar{n}_2$ (m^2/V^2) is the second order nonlinear refractive index and $|E^{in}|^2$ is electric field intensity inside the particles averaged over the nanoparticle volume. At both MD and ED resonances $|E^{in}|^2 \approx 10|E^{ext}|^2$, where $|E^{ext}|^2$ is the electric field intensity of the incident pulse. Peak intensity of the incident pulse is $I^{ext} = 2\varepsilon_0 c|E^{ext}|^2$. Commonly used nonlinear refractive index n_2 (m^2/W) is related to $\bar{n}_2$ through expression $n_2 = \bar{n}_2/(\varepsilon_0 n_L c)$[7]. From these relations we obtain $n = n_L + 10 n_2 n_L I^{ext}$.

Resonace frequency of the nanoparticle is approximately proportional to the value of refractive index, therefore relative shift of the resonance frequency ω_r as a function of the incident pulse intensity is expressed as following: $\delta = \dfrac{\Delta\omega_r}{\omega_r} = \dfrac{\Delta n}{n_L} = 10 n_2 I^{ext}$. In the main text we chose fixed value of nonlinear refractive index $n_2 = 8 \cdot 10^{-18}$ m^2/W, therefore e.g. for $I = 5$ GW/cm^2, we obtain $\delta = 0.4\%$ shift.

From equations (5) in linear regime we can obtain the absolute square of magnetic moment $\widetilde{M} = \dfrac{|\widetilde{H}^{(ext)}|^2}{\gamma_m^2}$ when a single particle is irradiated by a plane wave with normalized magnetic field $\widetilde{H}^{(ext)}$ at the MD resonance frequency. This is exactly the value of the relative resonance frequency shift δ which allow us to make correspondence between the $|\widetilde{H}^{(ext)}|^2$ in dimensionless units, used in calculations, and intensity of the incident pulse in W/m^2.

References

1. Mulholland, G. W.; Bohren, C. F.; Fuller, K. A. Light Scattering by Agglomerates: Coupled Electric and Magnetic Dipole Method. *Langmuir* **1994**, *10*, 2533–2546.

2. Merchiers, O.; Moreno, F.; González, F.; Saiz, J. M. Light scattering by an ensemble of interacting dipolar particles with both electric and magnetic polarizabilities. *Phys. Rev. A* **2007**, *76*, 043834.

3. Savelev, R. S.; Slobozhanyuk, A. P.; Miroshnichenko, A. E.; Kivshar, Y. S.; Belov, P. A. Subwavelength waveguides composed of dielectric nanoparticles. *Phys. Rev. B* **2014**, *89*, 035435.

4. Shore, R. A.; Yaghjian, A. D. Complex waves on periodic arrays of lossy and lossless permeable spheres: 1. Theory. *Radio Science* **2012**, *47*.

5. Evlyukhin, A. B.; Reinhardt, C.; Chichkov, B. N. Multipole light scattering by nonspherical nanoparticles in the discrete dipole approximation. *Phys. Rev. B* **2011**, *84*, 235429.

6. Noskov, R. E.; Belov, P. A.; Kivshar, Y. S. Subwavelength Modulational Instability and Plasmon Oscillons in Nanoparticle Arrays. *Phys. Rev. Lett.* **2012**, *108*, 093901.

7. Sutherland, R. L. *Handbook of nonlinear optics*; Marcel Dekker, New York, 1996.