QNMtoolbox_nonlinear: an openly available toolbox for nonlinear frequency-doubling nanooptics with quasinormal modes

Carlo Gigli[1], Tong Wu[2], Philippe Lalanne[2]

1. MPQ, Université de Paris
2. LP2N, Institut d'Optique d'Aquitaine, IOGS, Univ. Bordeaux, CNRS.

carlo.gigli@u-paris.fr

tong.wu@institutoptique.fr

philippe.lalanne@institutoptique.fr

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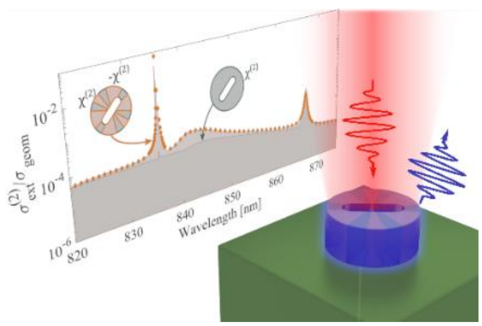

- the present user guide document,
- **QNMtoolbox_nonlinear.m**, a Matlab open-source script of the freeware package **MAN** (Modal Analysis of Nanoresonators) [1], built for extracting the normalized resonance modes (also called the quasinormal modes or QNMs) of dielectric nanoresonator [2], and solving nonlinear frequency-doubling problems in nanoresonators in the QNM basis. The script preferentially operates on the Matlab-COMSOL Livelink environment with the solver **QNMEig** of the package **MAN**; however, the code would also work with QNMs computed with other software,
- **QNMEig_NLnanodisk.mph**, an exemplary COMSOL model for operation with the **QNMEig** solver.The toolbox is distributed in the hope that it will be useful, but WITHOUT ANY WARRANTY; without even the implied warranty of MERCHANTABILITY or FITNESS FOR A PARTICULAR PURPOSE. See the GNU

General Public License for more details http://www.gnu.org/license/.
- a repertory Utilities composed of Matlab functions, to be included in the Matlab path. The functions in this folder should not be changed.

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1. INTRODUCTION TO QNMTOOLBOX_NONLINEAR

QNMtoolbox_nonlinear extracts the normalized QNMs of a nanoresonator on a substrate computed with the QNM solvers of the **MAN** package and computes the excitation coefficients associated to each QNM at real frequencies. The example is provided for a plane-wave excitation polarized along the x-axis propagating along the z-axis (TM polarization).

In this section, basic information about the main function of the toolbox **QNMtoolbox_nonlinear.m** is provided.

1.1 Download & Installation

QNMtoolbox_nonlinear.m is a Matlab script to be used with the Matlab-COMSOL livelink environment, in conjunction with the solver **QNMEig** and the model **QNMEig_NLnanodisk.mph**. The script needs to be placed in your Matlab folder to be run.

1.2 How to acknowledge and cite

We kindly ask that you reference the **MAN** package from IOGS-CNRS and its authors in any publication/report for which you used it.

- The preferred citation for **QNMtoolbox_nonlinear.m** is the following paper:
  [ref1] C. Gigli, T. Wu, G. Marino, A. Borne, G. Leo, and P. Lalanne, "Quasinormal-mode modeling and design in nonlinear nano-optics," 1–8 (2019).
- The preferred citation for **MAN** is the following paper:
  [ref2] W. Yan, R. Faggiani, and P. Lalanne, "Rigorous modal analysis of plasmonic resonators", Phys. Rev. B **97**, 205422 (2018).

A brief description of the algorithm might be:

"The nonlinear generation is computed in the QNM basis with QNMtoolbox_nonlinear [ref1], a toolbox of the freeware MAN (Modal Analysis of Resonators) [ref2] under the COMSOL Multiphysics environment."

1.3 Units and conventions of input/output data

Unit. All the input information is required to be in the SI unit. Accordingly, the output information is given in the SI unit as well.

Convention. The time dependent convention $\exp(i\omega t)$ used by COMSOL is adopted.

1.4 Outline of the theory and related key issues

QNMtoolbox_nonlinear.m is a Matlab script dedicated to the design of dielectric nanoresonators with nonlinear susceptibility. It relies on COMSOL Multiphysics®, its RF and Mathematics Modules, and MATLAB®.

Classically, nonlinear generation in nanoresonators is analyzed with two coupled Maxwell's equations to firstly determine the nonlinear local source generated inside the resonator induced by the external pump and then solve the radiation problem at the higher harmonic. This requires solving numerically Maxwell's equations twice, for the fundamental and the non-linear harmonic frequencies. The numerical load may be heavy, and, above all, the computed results may still hide a great deal of knowledge about the physical mechanisms at play. Modes represent a powerful characteristic of the resonator. If one is able to find these modes (they are called quasinormal modes) and understand how they are excited, then it is possible to describe how they are coupled through the nonlinear term.

The approach adopted by **QNMtoolbox_nonlinear.m** is exactly that one. It allows to compute numerically the QNMs of the resonator with a dispersive refractive index and to properly normalize them [2], to compute the excitation coefficients of these modes for any excitation pump with an analytical volume integration and finally to retrieve the excitation coefficients of the modes at the generation frequency [1]. For the sake of clarity, the toolbox focuses on the case of second harmonic generation in an AlGaAs nanocylinder, but it can be generalized to other nonlinear processes.

In more mathematical terms, we consider a plane wave $\mathbf{E}_{inc}(\mathbf{r},\omega)$ that is incident on the resonator. The the scattered field $\mathbf{E}_s(\mathbf{r},\omega,\mathbf{E}_b)$, defined as the difference between the fields in presence ($\mathbf{E}_{\mathrm{t}}(\mathbf{r},\omega)$) and absence ($\mathbf{E}_b(\mathbf{r},\omega)$) of the resonator, can be written with a modal expansion of the form

$$\mathbf{E}_s(\mathbf{r},\omega) = \sum_m \alpha_m^{(1)}(\omega)\,\tilde{\mathbf{E}}_m(\mathbf{r}), \tag{1}$$

where $\tilde{\mathbf{E}}_m$ denotes the electric-field map of the normalized QNM m with complex eigenfrequency $\tilde{\omega}_m$ and quality factor $Q = -\,\mathrm{Re}(\tilde{\omega}_m)\,/\,2\mathrm{Im}(\tilde{\omega}_m)$. The $\alpha_m^{(1)}$'s are the excitation coefficients that analytically depend on the background field. This implies that, once the resonant modes of the resonator are calculated, the optical response is known *analytically* (i.e. by numerical computation of simple overlap integrals between the background field $\mathbf{E}_b(\mathbf{r},\omega)$) for any frequency ω of the excitation field and the physical understanding is immediate and unambiguous since the mode expansion explicitly depends on the excitation parameters.

Numerical calculation of $\tilde{\mathbf{E}}_m$ can be performed with **QNMEig** (for instance with the model **QNMEig_NLnanodisk.mph** for the nanodisk) or **QNMPole**.

For dielectrics whose permittivity follows a single Lorentz pole model, $\varepsilon(\omega) = \varepsilon_\infty\left(1 - \frac{\omega_p^2}{\omega^2-\omega_0^2-i\gamma\omega}\right)$, the modal excitation coefficient can be written [2]

$$\alpha_m(\omega) = \left(\varepsilon_b - \varepsilon_\infty - (\varepsilon(\tilde{\omega}_m) - \varepsilon_b)\frac{\tilde{\omega}_m}{\tilde{\omega}_m-\omega}\right)\iiint_{V_{res}} \mathbf{E}_b\cdot\tilde{\mathbf{E}}_m d^3\mathbf{r}. \tag{2}$$

Note that if dispersion is not considered ($\omega_p = 0$), then Eq. (2) is reduced to

$$\alpha_m(\omega) = (\varepsilon_\infty - \varepsilon_{\mathrm{b}})\,\frac{\omega}{\tilde{\omega}_m-\omega}\iiint_{V_{res}} \mathbf{E}_b\cdot\tilde{\mathbf{E}}_m d^3\mathbf{r}. \tag{3}$$

The total field inside the resonator can be retrieved with several formulations, and consistently with convergence studies in [3], here we used

$$\mathbf{E}_{\mathrm{t}}(r,\omega) = \sum_m \alpha_m^{(1)}(\omega)\,\frac{\varepsilon(\tilde{\omega}_m)-\varepsilon_\infty}{\varepsilon(\omega)-\varepsilon_\infty}\,\tilde{\mathbf{E}}_m(\mathbf{r}). \tag{4}$$

Several studies – Tong Wu [3], Carlo Gigli [1] and Fenghao Xu (Brongersma group) – seem to consistently indicate that Eq. (4) is the formula that provides the fastest convergence rate. In the presence of a material with a second order nonlinear susceptibility $\chi^{(2)}$, the local current distribution generated inside the resonator volume is

$$\mathbf{J}^{(2)}(\boldsymbol{r},2\omega)=i2\omega\mathbf{P}^{(2)}(\boldsymbol{r},2\omega)=i2\omega\varepsilon_0\boldsymbol{\chi}^{(2)}:[\mathbf{E}_{\mathrm{t}}(\boldsymbol{r},\omega)\otimes\mathbf{E}_{\mathrm{t}}(\boldsymbol{r},\omega)],\qquad(5)$$

which is the source term of the Maxwell equation $\nabla\times\mathbf{H}_{\mathbf{t}}(\boldsymbol{r},2\omega)=i2\omega\varepsilon(\boldsymbol{r},2\omega)\mathbf{E}_{\mathbf{t}}(\boldsymbol{r},2\omega)+\mathbf{J}^{(2)}(\boldsymbol{r},2\omega)$. In analogy with equation (2), we define the excitation coefficients $\alpha_m^{(2)}(2\omega)$ at SH frequency

$$\alpha_l^{(2)}(2\omega)=-2\omega/(\tilde{\omega}_l-2\omega)\int_{V_r}\mathbf{P}^{(2)}(\boldsymbol{r},2\omega)\cdot\tilde{\mathbf{E}}_l(\boldsymbol{r})\,d\boldsymbol{r}.\qquad(6)$$

By analogy with (4), the total field at 2ω can be reconstructed with

$$\mathbf{E}_{\mathrm{t}}(2\omega)=\sum_{l=1}^{M_2}\alpha_l^{(2)}(2\omega)\tilde{\mathbf{E}}_l.\qquad(7)$$

Extinction and absorption cross sections at SH frequency are calculated using the QNM expansion

$$\sigma_{ext}^{(2)}(2\omega)=\frac{\mathrm{P_{ext}}(2\omega)}{S_0}=\frac{\omega}{S_0}\int_V Im\left[\sum_{l=1}^{M_2}\alpha_l^{(2)}(2\omega)\tilde{\mathbf{E}}_l(\boldsymbol{r})\cdot\mathbf{P}^{(2)^*}(\boldsymbol{r},2\omega)\right]d\boldsymbol{r},\qquad(8a)$$

$$\sigma_{abs}^{(2)}(2\omega)=\frac{P_{abs}(2\omega)}{S_0}=\frac{1}{2S_0}\int_V\frac{\gamma}{\varepsilon_\infty\omega_p^2}|\mathbf{J}(\mathbf{r},2\omega)|^2d^3\boldsymbol{r}\ ,\qquad(8b)$$

where S_0 is the time-averaged power of incident plane-wave at ω and $\mathbf{J}(\mathbf{r},2\omega)=\sum_l\alpha_l^{(2)}(2\omega)(i\tilde{\omega}_l(\varepsilon(\tilde{\omega}_l)-\varepsilon_\infty)\tilde{\boldsymbol{E}}_l(\boldsymbol{r}))$.

To summarize, the QNM computation and normalization (the prerequisite to using **QNMtoolbox_nonlinear.m**) are performed with **QNMEig**, using COMSOL software. The script extracts the normalized QNM fields inside the resonator domain, performs the overlap integral, and computes the modal excitation coefficients at pump frequency, reconstructs the field in the QNM basis. The present toolbox additionally computes also the nonlinear induced currents inside the resonator, the modal excitation coefficients and the reconstruction of total field at SH frequency, as well as the nonlinear cross sections.

Commenté [LP1]: This will be removed in future version.

2. SET-UP: EIGENMODES COMPUTATION IN COMSOL AND RECONSTRUCTION IN MATLAB

We recommend that the user starts with the nanorod example (QNMEig_Nanorod) that is provided to become familiar with QNM calculation and computation of excitation coefficient in absence of nonlinear generation effects before moving to the nonlinear generation analysis. To calculate and normalize QNMs, follow the following steps:

Commenté [LP2]: I think it is a recommendation that will rest even with the improved version of the software. See discussion with Carlo:
Yes why not, there should be a model for a non dispersive GaAs nanocylinder on the web. I left bowtie beacause I thought it was the exemplary tutorial to start with, but it is true that it is plasmonic and dispersive and it could just introduce unnecessary difficulties.

1/ Build on a COMSOL model sheet for your problem, or first use and modify the supplied model sheet for a AlGaAs nanodisk **QNMEig_NLnanodisk.mph**.

There are two equivalent alternatives to compute QNMs to study nonlinear generation in nanoresonators: The first one consists in searching eigenfrequencies around a real frequency at mid-way between fundamental frequency (FF) and second harmonic (SH) and ask COMSOL to find a large amount of modes. At variance, the second one consists in solving twice the same eigenvalue problem, centering the study first around FF and then around SH to reduce the number of desired modes to achieve satisfactory convergence. This userguide presents the second method due to computational costs advantages.

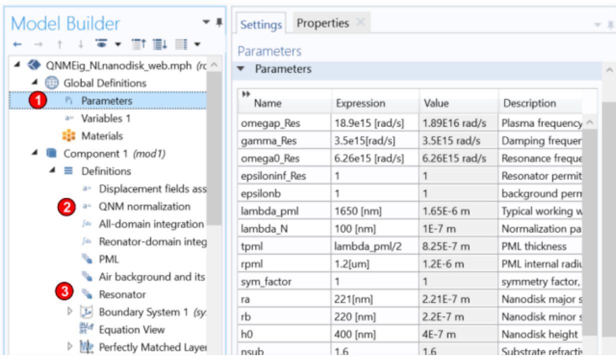

Figure 1. COMSOL Model QNMEig_NLnanodisk.mph interface.

In Parameters section under Global Definitions (see marker 1 in Fig.1) the user can find the following variables:

Parameter Name	Description
omegap_Res	Plasma frequency of single-pole Lorentzian used to model resonator permittivity
gamma_Res	Damping frequency of single-pole Lorentzian used to model resonator permittivity
omega0_Res	Resonance frequency of single-pole Lorentzian used to model resonator permittivity
epsiloninf_Res	Resonator permittivity at infinite real frequency
epsilonb	Background permittivity
lambda_pml	Working wavelength for which PML layers are optimized. This value can be centered between pumping wavelength and second harmonic
lambda_N	Normalization parameter used in the weak formulation.
tpml	Thickness of PML layer
rpml	PML internal radius. A spherical volume is considered in the model.
ra	Nanodisk basis major axis
rb	Nanodisk basis minor axis
h0	Nanodisk height
nsub	Substrate refractive index
z0	Background-substrate interface z-coordinate
lambda_target	Central working wavelength for eigenfrequency study. Study 1(QNM around FF) looks for eigenmodes around lambda_target while Study 2(QNM around SH) looks for eigenmodes around lambda_target_2

Study 1 (QNM around FF) and Study 2 (QNM around SH) compute field distributions (emw.Ex, emw.Ey, emw.Ez) and normalization factors QN (defined under "QNM normalization" variables context menu in "Definitions", see marker 2 in Fig.1) for eigenmodes around FF and SH respectively.

N.B: In order to visualize the normalized QNM profiles in COMSOL we recall that field distribution has to be divided by $\sqrt{QN}$ (e.g. emw.Ex/sqrt(QN), emw.normE/(abs(sqrt(QN))),…).

Commenté [LP3]: This will be automatically done in next versions.

2/ Open the Matlab script **QNMtoolbox_nonlinear.m**, and make sure that the COMSOL model in use is in the folder (see marker 1 in Fig.2).

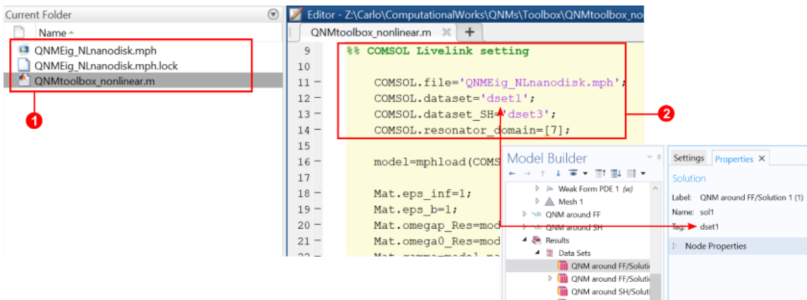

Figure 2. Matlab QNMtoolbox_nonlinear.m interface

3/ QNMtoolbox_nonlinear.m imports the QNM field distributions and eigenfrequencies from COMSOL and compute the reconstruction for a user-defined exciting field.
Matlab variables are classed in few structures as follows:

Structure	Field	Description
COMSOL	file	File name COMSOL model
	dataset	dataset tag solution QNM_FF study
	dataset_SH	dataset tag solution QNM_SH study
	resonator_domain	Domain label of resonator volume in COMSOL model
QNM/QNM_SH	omega	QNM complex frequencies $\tilde{\omega}_m$
	QN	QNM normalization coefficient
	cord	Evaluation points coordinates in resonator volume
	Ex/Ey/Ez	Electric field distribution x/y/z-component at coordinates "cord"
	mesh_vol	Volume of the discrete elements centered at coordinates "cord"
	eps	Material permittivity at frequencies "omega" $\varepsilon(\tilde{\omega}_m)$
Comp	lambda	Pump wavelengths simulation will be performed at
	I0	Pump intensity
	E0	Pump electric field absolute value
	phi	Pump linear polarization angle with respect to x-axis. $\phi = 0 \rightarrow \mathbf{E}_{\text{inc}} = E_0\hat{\boldsymbol{x}}$, $\phi = \pi/2 \rightarrow \mathbf{E}_{\text{inc}} = E_0\hat{\boldsymbol{y}}$
Sol/SolSH	alpha	Excitation coefficients $\alpha_m(\omega)$
	ext/extModes	Extinction cross section Total/Mode contributions
	abs/absModes	Absorption cross section Total/Mode contributions
	imax	Index of dominant modes

The script is divided in four sections:

- **COMSOL Livelink settings:** it defines the COMSOL structure to set-up the Livelink with QNMEigNLnanodisk.mph model (see marker 2 in Fig.2).
  In particular, please check the following parameters

- COMSOL.file: the name of the COMSOL file where the QNMs were computed.
- COMSOL.dataset: The tag of the data set "QNM around FF" solution.
- COMSOL.dataset_SH: The tag of the data set "QNM around SH" solution.
- COMSOL.resonator_domain: the index of the resonator domain (marker 3 in Fig.1).

- **Import modes from COMSOL:** it uses the COMSOL with Matlab Livelink to import field distributions and complex frequencies of modes around FF and SH.
- **Computational Settings:** in this section, settings for the reconstruction are defined: wavelength range, intensity and polarization of external excitation.
- **Computing Extinction, absorption cross section:** the section where excitation coefficients and cross sections are retrieved.

4/ Run the script. For every wavelength λ in Comp.lambda vector, $\alpha_m^{(1)}$ (Sol.alpha) and $\alpha_l^{(2)}$ (SolSH.alpha) will be computed for a plane wave excitation of amplitude Comp.E0 with k-vector along z-axis. Main script outputs the reconstructed cross sections at FF (Sol.ext, Sol.abs) and SH (SolSH.ext, SolSH.abs).

3. ADDITIONAL UTILITIES

QNM_toolbox_nonlinear offers few other additional functions in the folder "UTLILITIES" to analyze and plot the results of QNM reconstruction.

plot_QNMcomplexplane.m (QNM, Sol, Sol.alpha): visualize the frequencies of all the computed modes in the complex plane.

Field-distribution visualization (omega, nsol, model, COMSOL, Comp, SolSH, QNM_SH, CutPlane, varargin): reconstruct total nearfield at SH inside and outside the resonator. The inputs are listed as follows:

- omega: real frequency of pump excitation for which the reconstruction is performed
- nsol: indexes of the QNM used for reconstruction. The index of the first N dominant modes at SH are found in SolSH.imax
- model: the pointer to .mph model
- COMSOL: structure containing informations on COMSOL file
- Comp: structure containing information on the pump settings used for the simulation
- SolSH: structure containing the result of simulation at SH
- QNM_SH: structure containing QNM field distributions and eigenfrequencies
- CutPlane: plane where the modes are shown. It can assume the following values: 'XY', 'XZ' or 'YZ'
- Varargin: two optional input to define the region where the field will be shown. Varagin{1} defines the plot window in meters. For example if CutPlane='XY', varargin{1} can assume the form [x0,x1,y0,y1] meaning that the plot window will be restricted between x0 and x1 along x axis and between y0 and y1 along y axis. Varargin{2} defines the spatial resolution in meters.

compute_zeta (QNM, QNM_SH, Phy, Mat): compute the field overlap integral ζ_{lmn} defined in [1].

plot_OverlaTensor (zeta, imaxSH): show all the contributions to the SH mode with index l=imaxSH coming from all the FF modes.

4. Frequently asked questions

Which convention is used for plane waves?

COMSOL convention $\exp(i\omega t)$ is used. Note that the published article [1] uses the other convention.

How are the overlap integrals performed?

The values of the QNM fields are extracted for the coordinates contained in QNM.coord. The incident Electric field that overlaps with the QNM fields is defined analytically using the x, y, z coordinates in QNM.cord.

```
k=omega/Phy.c*sqrt(Mat.eps_b);        % wavenumber in background
p = exp(-1i*k*QNM.cord(1,:)*0-1i*k*QNM.cord(2,:)*0-1i*k*QNM.cord(3,:)); % Incident plane wave phase term
pr = exp(-1i*k*QNM.cord(1,:)*0-1i*k*QNM.cord(2,:)*0+1i*k*QNM.cord(3,:)); % Reflected plane wave at z=0 phase term
% Background field definition - valid for a plane wave with k vector along z axis
E_inc_x=(p+Comp.rs*pr).*Comp.E0*cos(Comp.phi); % Background electric field x-component
E_inc_y=(p+Comp.rs*pr).*Comp.E0*sin(Comp.phi); % Background electric field y-component
E_inc_z=p*0; % Background electric field z-component
```

The values of the overlap integral between a QNM mode and the incident field are stored inside the variable E_int :

```
% Field overlap integration between incident field and QNM field
  E_int=sym_factor.*sum( bsxfun(@times,QNM.Ex,E_inc_x.*QNM.mesh_vol)+...
        bsxfun(@times,QNM.Ey,E_inc_y.*QNM.mesh_vol)+...
        bsxfun(@times,QNM.Ez,E_inc_z.*QNM.mesh_vol),...
        2);
```

How are the modal excitation coefficients computed inside the program?

The prefactors to the modal excitation coefficients at frequency ω are stored inside the variable "alpha_QNM" which is defined as:

$$\text{alpha\_QNM} = \left(\varepsilon_b - \varepsilon_\infty - \left(\Delta\varepsilon(\tilde{\omega}_m)\right)\frac{\tilde{\omega}_m}{\tilde{\omega}_m - \omega}\right) = \frac{1}{\omega - \tilde{\omega}_m}\left(-\omega(\varepsilon_\infty - \varepsilon_b) - \tilde{\omega}_m\left(\varepsilon(\tilde{\omega}_m) - \varepsilon_\infty\right)\right). \quad (9)$$

The excitation coefficients are stored inside the structure Sol.alpha performing the following product:

```
Sol.alpha(:,iii) = (alpha_QNM.*E_int)./sqrt(QNM.QN); % Modal excitation coefficients
```

Factor $\sqrt{QN}$ at the denominator comes from the fact that QNM distributions inside E_int are not normalized.

How is the nonlinear current distribution computed?

According to Eq. (4) total field inside the resonator volume is reconstructed as it follows:

```
Et_x = sum (Sol.alpha(:,iii).*QNM.Ex./sqrt(QNM.QN).*(QNM.eps-Mat.eps_inf)/(Mat.eps(omega))- …
Mat.eps_inf)); % Total field at omega x-component
Et_y = sum (Sol.alpha(:,iii).*QNM.Ey./sqrt(QNM.QN).*(QNM.eps-Mat.eps_inf)/(Mat.eps(omega)- …
Mat.eps_inf)); % Total field at omega y-component
Et_z = sum(Sol.alpha(:,iii).*QNM.Ez./sqrt(QNM.QN).*(QNM.eps-Mat.eps_inf)/(Mat.eps(omega)- …
Mat.eps_inf)); % Total field at omega z-component
```

This field distribution is used to compute the second harmonic polarization vector, according to Eq. (5). This section is valid for second harmonic generation in zincblende materials and it has to be modified to extend the script to other processes in other materials.

```
PSH_x = 2*Phy.epsilon0*Mat.chi2*(Et_y.*Et_z); % Nonlinear polarization vector at SH x-component
PSH_y = 2*Phy.epsilon0*Mat.chi2*(Et_z.*Et_x); % Nonlinear polarization vector at SH y-component
PSH_z = 2*Phy.epsilon0*Mat.chi2*(Et_x.*Et_y); % Nonlinear polarization vector at SH z-component
```

The overlap integral $\int_{V_r} \boldsymbol{P}^{(2)}(\boldsymbol{r}, 2\omega) \cdot \widetilde{\boldsymbol{E}}_m(\boldsymbol{r})\, d\boldsymbol{r}$ in (6) is stocked inside the variable E_intSH:

```
E_intSH=sym_factor.*sum( bsxfun(@times,QNM_SH.Ex,PSH_x.*QNM.mesh_vol)+...
            bsxfun(@times,QNM_SH.Ey,PSH_y.*QNM.mesh_vol)+...
            bsxfun(@times,QNM_SH.Ez,PSH_z.*QNM.mesh_vol),...
            2);
```

5. References

[1] C. Gigli, T. Wu, G. Marino, A. Borne, G. Leo, and P. Lalanne https://arxiv.org/abs/1911.06373 (2019). "Quasinormal-mode modeling and design in nonlinear nano-optics"

[2] W. Yan, R. Faggiani, P. Lalanne, Phys. Rev. B **97**, 205422 (2018). "Rigorous modal analysis of plasmonic nanoresonators"

[3] T. Wu, A. Baron, P. Lalanne, and K. Vynck, Phys. Rev. A 011803, 1–5 (2020). "Intrinsic multipolar contents of nanoresonators for tailored scattering"