1. Introduction of the open-source numerical tool QNM-ED-LD for an efficient electrodynamics (ED) Langevin-dynamics (LD) simulation of multiple nanoparticles based on the coupling theory of quasinormal mode (QNM)

In reference [1] we propose an efficient method for simulating the motion of multiple nanoparticles under light-field excitation. In this method abbreviated as QNM-ED-LD, by exploiting the method proposed in reference [2] based on the coupling theory of quasinormal mode (QNM) [3] for an efficient electrodynamics (ED) calculation of optical force of multiple nanoparticles, one can achieve a fast solving of the Langevin dynamics (LD) equation which governs the motion of multiple nanoparticles and comprehensively incorporates the influence of optical force, viscous resistance, Brownian motion and the electrostatic double-layer force between particles. This method is generally applicable to any incident light beam and any geometries of nanoparticles. Compared with the ED-LD method based on full-wave finite element method (FEM) for calculating optical force (abbreviated as FEM-ED-LD), this method can reduce the computation time by at least two orders of magnitude. This method can be used as an efficient tool for designing optical tweezers with various applications.

In reference [1], as a numerical example, this method is used to simulate the motion of 40nm-radius spherical gold particles excited by a tightly focused laser beam of linear or radial polarization. In the simulation, gold particles move within a two-dimensional plane determined by the substrate surface. The refractive index of the substrate can be either the same as that of the water environment [as shown in Fig. 5(b1)] or different (as shown in Fig. S8, where the substrate is glass and the distance between the gold particles and the substrate is fixed). With minor modifications, this open-source numerical tool can be used for a fast simulation of the motion of particles in three-dimensional free space.

As typical numerical examples in reference [1], the codes for generating all curves in Fig. 5(b1) and Fig. S8 are provided in folders named "Fig5(b1)" and "FigS8", respectively, including the codes of QNM-ED-LD and FEM-ED-LD. The method for using the codes can be found in Section 3 "Instructions for Using the QNM-ED-LD Tool".

2. The softwares used by the QNM-ED-LD tool

The QNM-ED-LD open-source numerical tool uses COMSOL Multiphysics software (version 6.1) to calculate QNMs and the background field of focused laser beam, and uses MATLAB software (via COMSOL LiveLink for MATLAB, i.e. COMSOL with MATLAB) for electromagnetic-field and optical-force calculations as well as particle-motion simulations.

3. Instructions for Using the QNM-ED-LD Tool

3.1 Instructions for using the files in the folder 'Fig5(b1)'

Unpack the compressed file 'Fig5(b1).zip' to be the folder 'Fig5(b1)', where all relevant files are placed.

3.1.1 List of All Files

Table 3.1.1-1 Mph files (2)

Eb_rp_r40au_633_3np(0.95)_pz800.mph	Called by FEM-ED-LD main program “ausph_EDLD_3NP.m”, to calculate the electromagnetic field and optical force.
QNMEig_r40au_n1.33_24_1000nm_ewfd.mph	Solving QNMs.

Table 3.1.1-2 M files (12)

ausph_EDLD_3NP.m	QNM-ED-LD main program.
compare_EDLDresult_fig5.m	Displaying QNM-ED-LD and FEM-ED-LD simulation results.
comsol_EDLD3_auto_rp.m	FEM-ED-LD main program.
getQNM_save_to_mat_ewfd.m	Generating interpolation data files for the QNM fields.
electrostatics_Force.m	Subprogram called by the ED-LD main programs (including the QNM-ED-LD main program “ausph_EDLD_3NP.m” and the FEM-ED-LD main program “comsol_EDLD3_auto_rp.m”), to calculate the electrostatic double-layer force. It should be noted that this subprogram is only applicable to the situation described in reference [1]. When the particles or liquid environment changes, it should be considered whether the electrostatic double-layer force changes, and the calculation formula has been given in reference [1].
get_collision_dt.m	Subprogram called by the ED-LD main programs. When it is justified that particles experienced collision within a certain period of time, this program is used to obtain the accurate time of collision.
getaballweight.m	Subprogram called by the ED-LD main

	programs, to obtain the weight of a sphere (such as a gold nanosphere).
LD_beta.m	Subprogram called by the ED-LD main programs, to perform the LD calculation.
opticalForce.m	Subprogram called by the ED-LD main programs, to calculate optical force.
rand_vector_n.m	Subprogram called by the ED-LD main programs, to generate a three-dimensional random vector.
background_rp_B.m	Subprogram called by the ED-LD main programs, to calculate the magnetic-induction-intensity vector **B** of the tightly-focused radially-polarized laser beam.
background_rp_E.m	Subprogram called by the ED-LD main programs, to calculate the electric-field-intensity vector **E** of the tightly-focused radially-polarized laser beam.

3.1.2 Solving QNMs and generating data files

① In COMSOL Multiphysics software, open the file 'QNMEig_r40au_n1.33_24_1000nm_ewfd.mph', and click 'study 1' - 'compute' to solve QNMs. After completing the calculation, save the file and close the software. For the default parameters in reference [1], this calculation takes approximately 19 minutes. *For all the computation times mentioned in this document*, the computer configuration is the same as that in reference [1], namely a 4.6 GHz CPU and 64 GB RAM.

② In MATLAB software, run 'getQNM_save_to_mat_ewfd.m' to generate data files for the QNM fields. At this point, make sure that COMSOL with MATLAB is running. The program will generate 24 files in the folder 'Fig5(b1)' named as 'QNM_E_B_ewfdXX.mat', where XX is a number from 1 to 24, and each file (approximately 500-600 MB) corresponds to 1 QNM. The program will also generate a file named 'QNM_position_ewfd.mat' (approximately 5 MB). These files are the data files of QNM fields required by the QNM-ED-LD simulation program. In the data files, the coordinate range of the sampling points for the QNM field is $x \in [-1050,1050]$ nm, $y \in [-1050,1050]$ nm, $z \in [-50,50]$ nm, and the sampling interval is 4 nm. The coordinate origin is located at the center of the gold nanosphere. This selection of sampling points is a little overly dense for the simulation in Fig. 5 (b1) of reference [1], and the number of sampling points can be appropriately reduced to reduce the amount of data. This program will run for about 42 minutes.

3.1.3 QNM-ED-LD Simulation

3.1.3a QNM-ED-LD Simulation for the First Time

① Make sure that all the data files in the previous step of Section 3.1.2 have been generated.

② Make sure that COMSOL with MATLAB is running.

③ Make sure that 'readmat=0' is set in the program 'ausph_EDLD_3NP.m' (referred to as "Program 1" in this section).

④ Run Program 1. After running, the simulation results will be saved as 'OA3npb1.mat' by default.

Table 3.1.3a-1 Variables saved in 'OA3npb0.mat'

'position_x', 'position_y', 'position_z'	*x*, *y*, *z* coordinates of the center of each particle
'Fx', 'Fy', 'Fz'	*x*, *y*, *z* components of the optical force acting on each particle
'dertat'	A scalar variable, storing the identical step size for all time steps in the simulation.
'DERTAT'	A vector variable, storing the step sizes of every time steps in the simulation.
'POWER'	Power of the focused laser beam as the background field, in unit of mW.
'brown'	Control variable for the Brownian motion.
'flag_elec'	Control variable for the electrostatic double-layer force.

Note 1: In Program 1, 'brown=1' or 'brown=0' means that the Brownian motion is considered or not, respectively.

Note 2: In Program 1, 'flag_elec=1' or 'flag_elec=0' means that the electrostatic double-layer force is considered or not, respectively. The 'electrostatics_Force.m' called by Program 1 is used to calculate the electrostatic double-layer force between particles. If the particles change, please modify the 'electrostatics_Force.m' according to the relevant description in reference [1].

Note 3: After Program 1 completes, a file named 'QNM_qiu_1000nm_glass_d15_sel15_ewfd.mat' (2.8 MB) will be generated. This file contains the mesh data of the QNM field on the closed surface *S* surrounding the particle, which is used for calculating the optical force (including mesh point coordinates, mesh vectors, etc., but not the QNM field itself). If the QNMs are not changed afterward, the following actions can be performed:

(1) Change 'readmat=0;' to be 'readmat=1;' in Program 1.

(2) Do not need to run COMSOL with MATLAB, and just run Program 1 in MATLAB to complete the simulation.

3.1.3b QNM-ED-LD Simulation for the Second and Subsequent Times

As stated in Note 3 of Section 3.1.3a ④, if the QNMs are not changed, there is no need to run COMSOL with MATLAB. You can complete the simulation simply by running the program 'ausph_EDLD_3NP.m' in MATLAB.

3.1.4 FEM-ED-LD Simulation

① Make sure that COMSOL with MATLAB is running.

② Run the program 'comsol_EDLD3_auto_rp.m', where 'Eb_rp_r40au_633_3np(0.95)_pz800.mph' is called. After the program is completed, manually determine whether to terminate the simulation. If you wish to continue the simulation, assign the completed number of time steps of simulation to the variable 'kkk' and assign the number of time steps for the continued simulation to the variable 'num_n'. Then, rerun the program. The program saves results at each time step, thus allowing the simulation to be terminated at any point. The simulation results are saved by default to 'comsol_r40au_of_rp_0.95n1.33_633_z800_m20_edld_3np_binding_auto.mat'.

Note 1: Each time step requires the calculation of the background field of focused laser beam. Because the background field is defined by using a formula rather than interpolation from existing data, the calculation of electromagnetic field in COMSOL is slow and takes several minutes for one time step. So please be patient. If the background field is pre-calculated and saved as an interpolation function in 'Eb_rp_r40au_633_3np(0.95)_pz800.mph', the computation time can be reduced to about 1/3 (please refer to the FEM-ED-LD program 'Eb_rp_r40au_633_3np(0.95)_pz800_glass.mph' in the folder 'FigS8').

3.1.5 Displaying QNM-ED-LD and FEM-ED-LD simulation results

Run the program 'compare_EDLDresult_fig5.m', which can directly display all the curves in Fig. 5(b1) of reference [1].

3.2 Instructions for using the files in the folder 'FigS8'

Unpack the compressed file 'FigS8.zip' to be the folder 'FigS8', where all relevant files are placed.

3.2.1 List of All Files

Table 3.2.1-1 Mph files (4)

background_withglass_port_2D.mph	Calculating the electromagnetic field for a tightly-focused radially-polarized laser beam which irradiates a glass

	substrate, to obtain a two-dimensional distribution of field on the cross section y=0, which is used to generate a three-dimensional distribution of field in space.
Eb_rp_r40au_633_2np(0.95)_pz800_portglass_BACKground_comport_ewfd.mph	Calculating the electromagnetic field for a tightly-focused radially-polarized laser beam which irradiates a glass substrate, to obtain a three-dimensional distribution of field in space.
Eb_rp_r40au_633_3np(0.95)_pz800_glass.mph	Called by FEM-ED-LD main program "comsol_EDLD3_auto_rp_glass.m", to calculate the electromagnetic field and optical force.
QNMEig_r40auglass_n1.33_24_1000nm_ewfd.mph	Solving QNMs.

Table 3.2.1-2 M files (11)

ausph_EDLD_3NP_glass.m	QNM-ED-LD main program.
compare_EDLDresult_figs8.m	Displaying QNM-ED-LD and FEM-ED-LD simulation results.
comsol_EDLD3_auto_rp_glass.m	FEM-ED-LD main program.
getbackground_save_to_mat_comport_ewfd.m	Generating interpolation data file for the background field.
getQNM_save_to_mat_ewfd_glass.m	Generating interpolation data files for the QNM fields.
electrostatics_Force.m	Subprogram called by the ED-LD main programs (including the QNM-ED-LD main program "ausph_EDLD_3NP_glass.m" and the FEM-ED-LD main program "comsol_EDLD3_auto_rp_glass.m"), to calculate the electrostatic double-layer force. It should be noted that this subprogram is only applicable to the situation described in reference [1]. When the particles or liquid environment changes, it should be considered whether the electrostatic double-layer force changes, and the calculation formula has been given in reference [1].
get_collision_dt.m	Subprogram called by the ED-LD main programs. When it is justified that particles experienced collision within a certain period of time, this program is used to obtain the accurate time of collision.

getaballweight.m	Subprogram called by the ED-LD main programs, to obtain the weight of a sphere (such as a gold nanosphere).
LD_beta.m	Subprogram called by the ED-LD main programs, to perform the LD calculation.
opticalForce.m	Subprogram called by the ED-LD main programs, to calculate optical force.
rand_vector_n.m	Subprogram called by the ED-LD main programs, to generate a three-dimensional random vector.

3.2.2 Solving QNMs and generating data files

① In COMSOL Multiphysics software, open the file 'QNMEig_r40auglass_n1.33_24_1000nm_ewfd.mph', and click 'study 1' - 'compute' to solve QNMs. After completing the calculation, save the file and close the software. For the default parameters in reference [1], this calculation takes approximately 25 minutes.

② In MATLAB software, run 'getQNM_save_to_mat_ewfd_glass.m' to generate data files for the QNM fields. At this point, make sure that COMSOL with MATLAB is running. The program will generate 24 files in the folder 'FigS8' named as 'QNM_E_B_glass_ewfdXX.mat', where XX is a number from 1 to 24, and each file (approximately 500-600 MB) corresponds to 1 QNM. The program will also generate a file named 'QNM_position_glass_ewfd.mat' (approximately 5 MB). These files are the data files of QNM fields required by the QNM-ED-LD simulation program. In the data files, the coordinate range of the sampling points for the QNM field is $x\in[-1050,1050]$ nm, $y\in[-1050,1050]$ nm, $z\in[-50,50]$ nm, and the sampling interval is 4 nm. The coordinate origin is located at the center of the gold nanosphere. This selection of sampling points is a little overly dense for the simulation in Fig. S8 of reference [1], and the number of sampling points can be appropriately reduced to reduce the amount of data. This program will run for about 42 minutes.

3.2.3 Solving the background field and generating data files

① Open 'background_withglass_port_2D.mph', and click 'study 1' - 'compute' to calculate the background field. After completing the calculation, click 'Results' - 'Export' - 'data 1', In the menu bar 'Output', set the folder for exporting files to be 'FigS8' and the file name to be "filename" (which can be any name), and select the file format as 'txt'. Other settings have been set and do not need to be changed. Click the 'Export' button in the 'Settings' submenu to export the calculation results. Save and close the program. For the default parameters of the program (grid set as the maximum of 20 nm within and near the region of ED-LD simulation, and grid set as the maximum of 100 nm far away from the region of ED-LD simulation), the program will run for about 1 minute.

② Open 'Eb_rp_r40au_633_2np(0.95)_pz800_portglass_BACKground_comport_ewfd.mph'.

(1) Click 'Component 1' - 'Definitions' - 'E_interpolation', and select 'File' as the data source.
(2) Click 'Browse' in the 'Filename' submenu and select the txt data file generated in step ①.
(3) Set the 'Data format' to be 'spreadsheet' and the 'number of arguments' to be '2'.
(4) Click 'import' in the 'Filename' submenu.
(5) Click 'study 1' - 'compute'. After completing the calculation, save the file and close the software. For the default parameters of the program (selecting 'Physics-controlled mesh' in all calculation regions), this program will run for about 5 minutes.

③ Run 'getbackground_save_to_mat_comport_ewfd.m'. At this point, make sure that COMSOL with MATLAB is running. The program will generate 2 files in the folder 'FigS8': 'BACKground_glass_gap20_comport_ewfd.mat' (825 MB) and 'BG_position_glass_comport_ewfd.mat' (4.3 MB), which are the background-field data files required for QNM-ED-LD simulation. In the data files, the coordinate range of the sampling points for the background field is $x\in[-1200,1200]$ nm, $y\in[-1200,1200]$ nm, $z\in[-50,50]$ nm, and the sampling interval is 4 nm. The coordinate origin is set so that if there was no glass substrate, the focal point is at $(x, y, z)=(0, 0, -800)$ nm. This selection of sampling points is a little overly dense for the simulation in Fig. S8 of reference [1], and the number of sampling points can be appropriately reduced to reduce the amount of data. This program will run for about 1.7 minutes.

④ Open 'Eb_rp_r40au_633_3np(0.95)_pz800_glass.mph'.

(1) Click 'Component 1' - 'Definitions' - 'E_interpolation', and select 'File' as the data source.
(2) Click 'Browse' in the 'Filename' submenu and select the txt data file generated in step ①.
(3) Set the 'Data format' to be 'spreadsheet' and the 'number of arguments' to be '2'.
(4) Click 'import' in the 'Filename' submenu.
(5) Save the file and close the COMSOL Multiphysics software.

3.2.4 QNM-ED-LD Simulation

3.2.4a QNM-ED-LD Simulation for the First Time

① Make sure that all the data files in the previous steps of Sections 3.2.2 and 3.2.3 have been generated.

② Make sure that COMSOL with MATLAB is running.

③ Make sure that 'readmat=0' is set in the program 'ausph_EDLD_3NP_glass.m' (referred to as "Program 1" in this section).

④ Run Program 1. After running, the simulation results will be saved as 'OA3npglassb1.mat' by default.

Table 3.2.4a-1 Variables saved in 'OA3npglassb1.mat'

'position_x', 'position_y', 'position_z'	x, y, z coordinates of the center of each particle
'Fx', 'Fy', 'Fz'	x, y, z components of the optical force acting on each particle
'dertat'	A scalar variable, storing the identical step size for all time steps in the simulation.
'DERTAT'	A vector variable, storing the step sizes of every time steps in the simulation.
'POWER'	Power of the focused laser beam as the background field, in unit of mW.
'brown'	Control variable for the Brownian motion.
'flag_elec'	Control variable for the electrostatic double-layer force.

Note 1: In Program 1, 'brown=1' or 'brown=0' means that the Brownian motion is considered or not, respectively.

Note 2: In Program 1, 'flag_elec=1' or 'flag_elec=0' means that the electrostatic double-layer force is considered or not, respectively. The 'electrostatics_Force.m' called by Program 1 is used to calculate the electrostatic double-layer force between particles. If the particles change, please modify the 'electrostatics_Force.m' according to the relevant description in reference [1].

Note 3: After Program 1 completes, a file named 'QNM_qiu_1000nm_glass_d15_sel15_ewfd.mat' (2.8 MB) will be generated. This file contains the mesh data of the QNM field on the closed surface S surrounding the particle, which is used for calculating the optical force (including mesh point coordinates, mesh vectors, etc., but not the QNM field itself). If the QNMs are not changed afterward, the following actions can be performed:

(1) Change 'readmat=0;' to be 'readmat=1;' in Program 1.

(2) Do not need to run COMSOL with MATLAB, and just run Program 1 in MATLAB to complete the simulation.

3.2.4b QNM-ED-LD Simulation for the Second and Subsequent Times

As stated in Note 3 of Section 3.2.4a ④, if the QNMs are not changed, there is no need to run COMSOL with MATLAB. You can complete the simulation simply by running the program 'ausph_EDLD_3NP_glass.m' in MATLAB.

3.2.5 FEM-ED-LD Simulation

① Make sure that COMSOL with MATLAB is running.

② Run the program 'comsol_EDLD3_auto_rp_glass.m', where 'Eb_rp_r40au_633_3np(0.95)_pz800_glass.mph' is called. After the program is completed, manually determine whether to terminate the simulation. If you wish to continue the simulation, assign the completed number of time steps of simulation to the variable 'kkk' and assign the number of time steps for the continued simulation to the variable 'num_n'. Then, rerun the program. The program saves results at each time step, thus allowing the simulation to be terminated at any point. The simulation results are saved by default to 'comsol_r40au_of_rp_0.95n1.33_633_z800_m24_edld_3np_glass_binding_auto.mat'.

3.2.6 Displaying QNM-ED-LD and FEM-ED-LD simulation results

Run the program 'compare_EDLDresult_figs8.m', which displays Figs. S8 (a), (b) and (c) in reference [1] by assigning the variable 'flag' = 1, 2 and 3, respectively.

It should be noted whether the data of each curve in Fig. S8 of reference [1] considers Brownian motion. For example, in Fig. S8 (b) of reference [1], the QNM-ED-LD data does not consider the Brownian motion. If the QNM-ED-LD data considering Brownian motion is mistakenly read, the generated curve will be different from that in Fig. S8 (b) of reference [1].

4. Citing QNM-ED-LD

We kindly request that you cite the following Refs. [1-3] in any published work for which you used our provided codes here.

5. References

[1] Lu, X.; Tao, Q.; Zhong, Y.; Liu, H. Efficient method for the electrodynamics Langevin-dynamics simulation of multiple nanoparticles based on the coupling theory of quasinormal mode. ACS Photonics 2024, 11 (8), 2970-2980.

[2] Qi, Z.; Zhong, Y.; Liu, H. Efficient method for the calculation of the optical force of multiple nanoparticles based on the coupling theory of quasinormal modes. Opt. Lett. 2021, 46 (18), 4610-4613.

[3] Tao, C.; Zhu, J.; Zhong, Y.; Liu, H. Coupling theory of quasinormal modes for lossy and dispersive plasmonic nanoresonators. Phys. Rev. B 2020, 102 (4), 045430.