

Simulating electron energy-loss spectroscopy and cathodoluminescence for nanoparticles located on substrates

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Background

Electron energy-loss spectroscopy (EELS) and cathodoluminescence (CL) are widely used to study the plasmonic properties of individual nanoparticles. In the experiment, the sample is irradiated with relativistic electrons having energy of about 100 keV. After the interaction with the sample, electrons' energies are measured to obtain the energy loss spectrum (EELS spectrum). When interacting, the sample itself begins to emit photons. This light is collected by a parabolic mirror to obtain CL spectrum. The main advantage of EELS/CL over light scattering methods is the ability to study plasmonic properties with a spatial resolution of less than 1 nm (at wavelengths of visible light and infra-red).

Methods

We extended the capabilities of the discrete dipole approximation (DDA) to simulate EELS and CL. Until recently, there was only a theory for simulating the case of particles located in vacuum, which is never true in the experiment. In an electron microscope, the particle is always placed inside or on top of the substrate medium, which redshifts the plasmonic response of the sample. We extended our previously published DDA theory for particles inside an infinite medium[1] to simulate EELS and CL for particles on top of a semi-infinite medium, which successfully reproduces experimental results for particles deposited on TEM membranes of finite thickness.

Results

With the extended theory, the simulations successfully reproduce the results of EELS experiment where a gold nanoprisim (209 nm side length, 10 nm thickness) was deposited on 30-nm-thick Si₃N₄ substrate.[2] A straightforward workaround for such simulations (with any DDA code) is to discretize a finite volume of the substrate below the particle, which would take 1 645 070 dipoles for both prism and the substrate (Fig. 1a), making it a heavy computational task. In the extended theory, however, the presence of substrate is accounted in the DDA formulation of the problem (by adjusting the dipole-dipole interaction function) requiring only 66 206 dipoles to discretize only the prism itself (Fig. 1b). This requires much less computational resources, leading to about 10 times faster simulations.

In the simulations of the prism, the Si₃N₄ refractive index was set to a constant 2.15 for all energies, and the refractive index of gold was taken from the data by Babar et al.[3] The redshift caused by the semi-infinite substrate in the simulated plasmon energy peaks matches the ones observed in the experiment by Griffin et al.[2]. Moreover, the simulated plasmon maps (Fig. 1c) also match the experimental ones,[2] in contrast to the simulations for a particle in vacuum in that paper.

The extended theory also successfully reproduces the CL experiment where a gold nanorod was placed on top of SiO₂ membrane,[4] and even the CL experiment where a gold nanorod-shaped antenna was on top of gold substrate.[5]

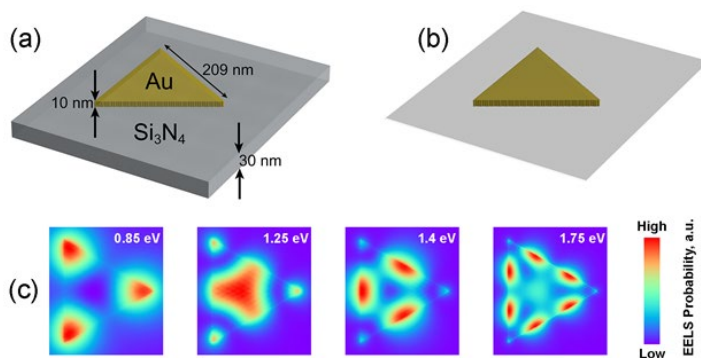
Conclusion

Simulations with the semi-infinite substrate theory successfully reproduce experimental spectra and plasmon maps for both EELS and CL for samples on top of dielectric substrates, and for CL even in the case of a metallic substrate. Brief theory overview followed by simulation results in comparison with previously published and our own experiments will be presented at the conference.

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Graphic:



Keywords:

Plasmonics, EELS, cathodoluminescence, DDA

Reference:

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