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Abstract: We perform simulations to model electron energy loss spectroscopy of aluminum nanodisk surface plasmons. Nanodisk geometry and e-beam position determine excitation and energy of plasmonic modes. Multipolar modes are explained with a circulating waveguide model.

OCIS codes: (240.6680) Surface plasmons; (310.6628) Subwavelength structures, nanostructures

Aluminum (Al) nanostructures support high energy plasmonic resonances at visible to ultraviolent (UV) and deep-ultraviolet (DUV) wavelengths [1, 2], due to low optical loss at these wavelengths and a high plasmon energy of Al. Moreover, Al is cheap, naturally abundant, and CMOS-compatible. These properties make Al a promising candidate plasmonic material for large scale manufacturing and commercial applications. Meanwhile, electron energy loss spectroscopy (EELS) has been used to probe plasmonic nanostructures with a high spatial resolution and the ability to resolve non-radiative, dark plasmon modes [2, 3]. There has also been an increasing effort in understanding the theory behind EELS investigation of plasmonic/photonic modes and implementing numerical simulations to study the interactions between an electron beam and plasmonic/photonic structures [4, 5].

Here we use numerical simulations to study EELS of Al nanodisks with 20 nm thickness and 20-120 nm diameter. Numerical calculations are performed in *COMSOL Multiphysics*. Al nanodisks are placed on a 5-nm-thick SiN_x membrane. We also include a 2.6-nm-thick native aluminum oxide layer. The optical constants of materials are taken from experimental data. The electron beam is modeled as a current pulse induced by a linearly moving charge. Multipolar surface plasmon modes as well as center-symmetric plasmonic breathing modes are revealed, with resonant energies ranging from 2 eV to 8 eV (Fig. 1a). Most plasmonic modes are blueshifted with a decreasing nanodisk diameter, with a few exceptions which are identified as surface plasmon modes at Al/SiN_x interface.

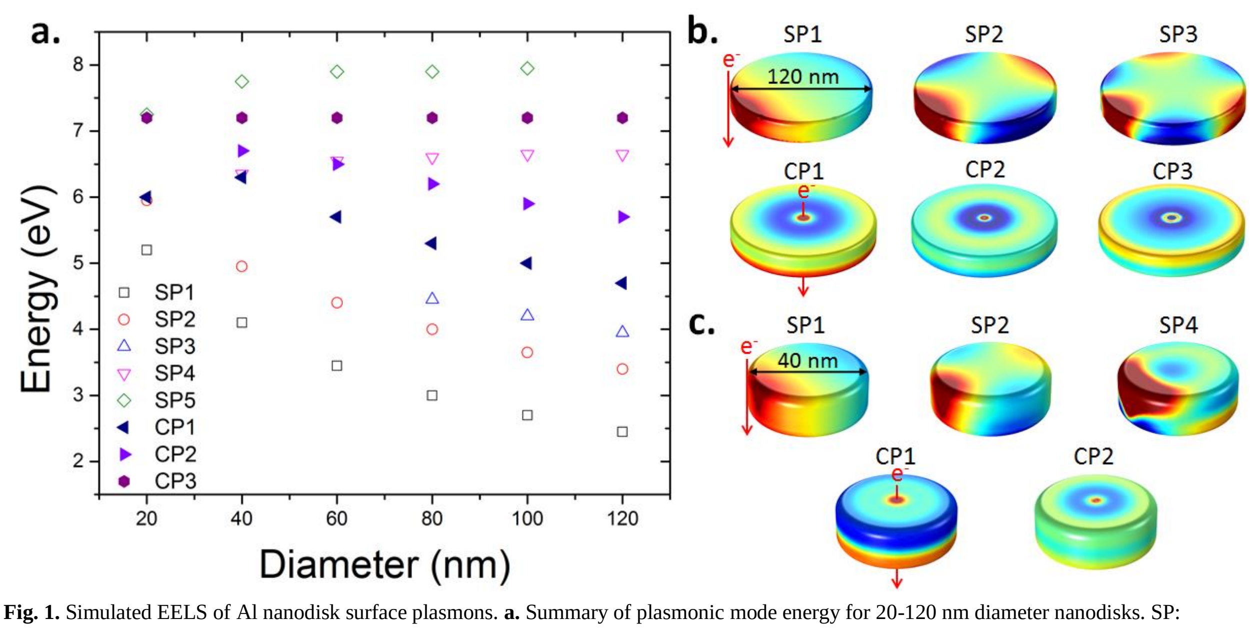

Fig. 1. Simulated EELS of Al nanodisk surface plasmons. **a.** Summary of plasmonic mode energy for 20-120 nm diameter nanodisks. SP: "surface plasmon", excited with the electron-beam at the edge of the nanodisk; CP: "center plasmon", excited with the electron-beam at the center of the nanodisk. **b.** & **c.** Surface normal electric field maps for SP and CP modes in nanodisks with 120 nm and 40 nm diameters. The excitation electron beam is shown as a red arrow in the maps for the first SP and CP modes of each diameter.

The excitation intensity of different modes depends on electron-beam position, as well as nanodisk geometry (Fig. 1b,c). When the electron beam is positioned at the edge of the nanodisk, the excited plasmonic modes are referred to as "surface plasmon" or SP modes. SP modes are predominantly multipolar modes. Fig. 1b shows SP1, SP2, and SP3 modes of a 120 nm diameter Al nanodisk are dipole, quadrupole, and hexapole modes, respectively. When the electron beam is positioned at the center of the nanodisk, the excited plasmonic modes are referred to as "center plasmon" or CP modes. CP modes are center-symmetric plasmonic breathing modes, as can be seen in Fig. 1b. Interestingly, when the nanodisk diameter gets smaller, the excitation of multipolar modes and center-symmetric breathing modes are no longer solely dependent on the electron beam position. For example, Fig. 1c shows SP and CP modes of a 40 nm diameter Al nanodisk. The SP4 mode, even though excited by an electron beam at the edge of the nanodisk, is essentially a center-symmetric breathing mode, as can be seen from the surface field map as well as its resonant energy, which coincides with the energy of CP1 mode.

We also explain the multipolar modes with a simple yet effective circulating plasmonic waveguide model. The waveguide is formed at the edge of the nanodisk. The surface plasmon wave in the waveguide travels along the sidewall and circulates the nanodisk. The dispersion relation, as well as electric and magnetic field profiles of the waveguide mode are illustrated in Fig 2, obtained from the 2D model solver in *COMSOL*. Fig. 2a also shows the nanodisk multipolar modes obtained in the EELS simulation in the energy-momentum space, with the momentum calculated by assuming the multipolar modes form standing wave patterns at the nanodisk perimeter. Close fitting between the plasmonic waveguide mode and the nanodisk multipolar modes indicates the multipolar modes can be well explained by the simple circulating waveguide model. The 2D simulation reproduces 3D simulation results with a reasonable accuracy and much less computation.

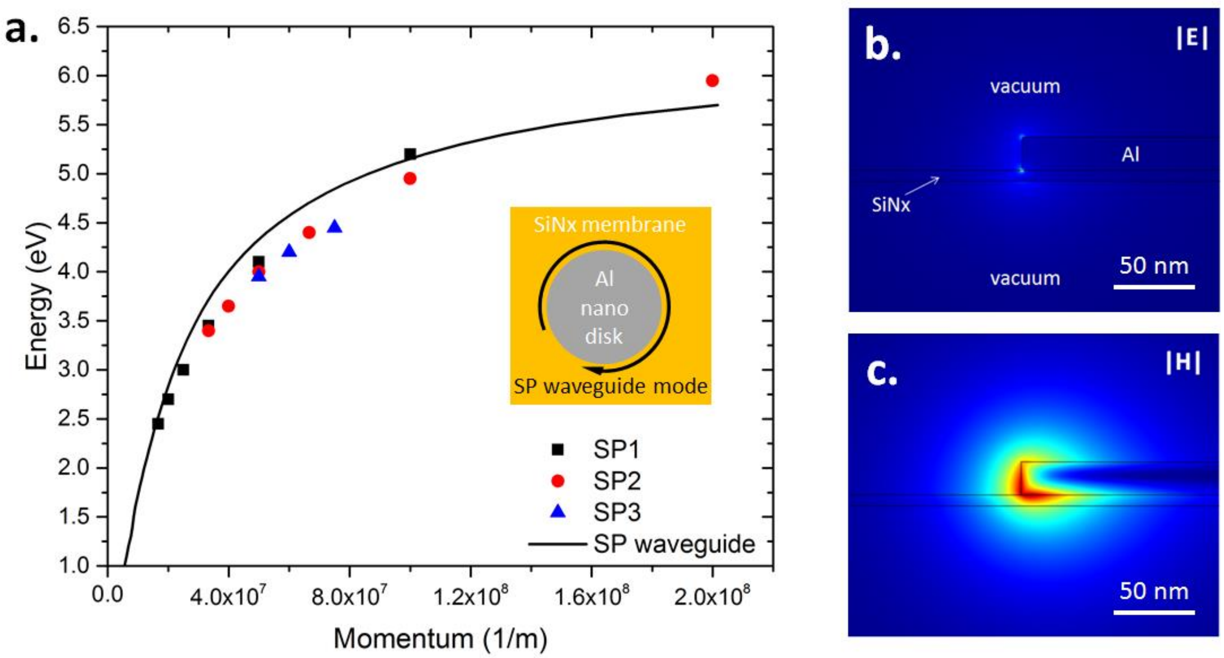

Fig. 2. Multipolar plasmonic modes explained by a simple circulating plasmonic waveguide model. **a.** Dispersion relation of the circulating surface plasmon waveguide mode, and the nanodisk multipolar modes in the energy-momentum space. The momentum of the multipolar modes is calculated by assuming these modes form a standing wave at the perimeter of the nanodisk. Inset: a top-view schematic of the nanodisk-on-a-membrane structure; the black curved arrow indicates the propagation of the circulating plasmonic waveguide mode. **b.** & **c.** |E| and |H| fields of the plasmonic waveguide mode obtained from the 2D mode solver. The waveguide geometry emulates the cross-section of the nanodisk-on-a-membrane structure.

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