Abstract

Strong light-matter coupling offers the possibility of altering material properties at the fundamental level. In recent years, the strong coupling phenomena have gained wide attention across many fields. Traditionally, strong coupling has been achieved using external optical cavities, which offer flexibility in selecting interacting components. However, this approach has limitations, such as requiring sophisticated fabrication techniques and complex experimental setups, which restrict scalability and practical applications. A compelling approach is self-hybridization, which allows optical modes and excitonic transitions to exist together in the same material, thereby avoiding the need for external cavity structures. Self-hybridization simplifies fabrication while maintaining the advantages of strong coupling, leading to efficient and compact device architectures.

This dissertation examines strong coupling between optical modes and excitons in self-hybridized uniaxial hyperbolic multilayer nanospheroids, systematically analyzing the effects of geometry and shape on these coupling phenomena. The nanospheroids have a wide range of aspect ratios that include prolate, sphere, and oblate geometries. Numerical and computational methods used for this study include the T-matrix and finite difference time domain (FDTD) techniques.

The first part of the research is the study of the optical modes of uniaxial hyperbolic nanospheriods that are composed of silver/silica multilayers. Their optical response and mode coupling characteristics are investigated in relation to their shape as the aspect ratio is varied from 1/3 (prolate) to 1 (sphere) to 3 (oblate). The optical responses are dependent on the material anisotropy, shape, and type of illumination. The results show that two dominant modes are present in the system: an electric dipole mode (ED) coupled to a magnetic quadrupole (MQ) and a magnetic dipole mode (MD) coupled to an electric quadrupole (EQ). The oblate shape exhibits less favorable optical mode characteristics compared to the prolate and spherical shapes.

Building upon these results, the second part of the research encompasses self-hybridized strong coupling of the optical modes to excitons in prolate and spherical geometries. Here, excitons are embedded within the material matrix as a function of their optical properties. The prolate and spherical nanoparticles exhibit large Rabi splitting as the oscillator strength is varied from 0 to 1 enabling strong coupling of the optical modes (ED and MD) to the excitons. The results show under which conditions the MD or ED modes offer greater coupling to excitonic or molecular transitions in the prolate and spherical geometries.

Building upon the MD dominance, the third part of the research focuses on evaluating practical realization of self-hybridized strong coupling of MD modes to excitons in the prolate and spherical geometries using TMDC (transition metal dichalcogenide) materials like MoS_2 (molybdenum disulfide), $MoSe_2$ (molybdenum diselenide), WS_2 (tungsten disulfide), and WSe_2 (tungsten diselenide). First, general considerations of coupling TMDCs to modes of hyperbolic nanoparticles are evaluated for simple two-component structures, which, however, yield an absence of strong coupling due to spectral detuning of the modes. To solve this limitation, a dielectric spacer is introduced to make the system a three-layered nanospheroid (silver/silica/TMDCs). The results show that the strong coupling in the MD regime is achieved in the three-layered prolate geometry composed of silver/silica/WS_2 and silver/silica/MoS_2.

In summary, this dissertation provides insight into how geometry and shape influence or suppress strong coupling in uniaxial hyperbolic multilayer nanospheroids. These can enhance the design of the next generation of opto-electronics devices.