

PhD THESIS DEFENSE: Nonlocal and nonlinear effects in nanophotonics

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The fundamental science and technological applications of light-matter interactions on nanometer length scales form the field of study known as nanophotonics. Explorations in nanophotonics expand our understanding at the interface between classical and quantum physics, while offering the means to address key societal challenges presented by the information and communication age, particularly concerning the development of light-based technologies that perform faster and more efficiently than their electronic counterparts. Light consists of propagating electromagnetic waves that can be guided, diffracted, or scattered through their interaction with matter. As a wave, light is characterized by its wavelength in free space, with the visible spectrum corresponding to ~400 - 800 nm, while the infrared and ultraviolet regimes of electromagnetic radiation emerge at wavelengths just above and below the visible range, respectively. To control light on the nanoscale, one must

overcome the well-known Abbe limit of diffraction that prevents the focusing of light on length scales below the optical wavelength, which can be circumvented by employing optical resonances in materials. In particular, we explore plasmons-the collective oscillations of conduction electrons in metals-as a platform to concentrate electromagnetic energy down to nanometric volumes, enhancing the associated electromagnetic fields and light-matter interactions. Noble metals such as gold, silver, and copper represent the standard choice of material in the study of subwavelength optics and plasmonics, while recent advancements in nanofabrication enable customization of their plasmon resonances. In this thesis, we theoretically explore the interaction of light (comprising the mid-infrared, visible, and UV parts of the electromagnetic spectrum) with noble metal films engineered with atomic-scale precision.

The manuscript starts with a comprehensive introduction of plasmons in metallic films, emphasizing the unique features that make these subwavelength optical excitations appealing for implementation in technological applications. We go beyond classical electromagnetism approaches by incorporating semi-classical models to describe the optical response of matter at the atomic level, which involves further complexity. Accordingly, the first and second chapters of the thesis are devoted to the introduction of classical and quantum mechanical descriptions of plasmons in metallic films, respectively. Chapter 3 utilizes these pillars as a foundation to study three aspects of light-matter interactions in metallic thin films at the nanoscale on which we concentrate: surface effects, interaction with electron beams, and heterostructure architectures in which we combine ultra-thin metallic films with two-dimensional materials such as graphene.

Once we have analyzed different aspects of the linear properties of the plasmonic response, Chapter 4 focuses on the nonlinear optical behaviour of metal films. The first part describes the intrinsic nonlinear properties of metal films, whereas the second part explores a specific nonlinear phenomenon: two-photon luminescence in gold films. Following up with nonlinear properties but deviating from the use of metallic thin films, we propose in Chapter 5 a path to channel entangled photons encoded in the optical modes of a waveguide and excite them by direct external illumination, leveraging the nonlinear properties of the waveguide and that overcomes involved optical elements commonly used in the generation of entangled photon pairs.

Overall, the thesis introduces a quantum mechanical model to understand the plasmonic properties of noble metal films, revealing the benefits of few-atom-thick films for boosting light-matter interaction at the nanoscale. We envision that our findings contribute to broadening the fundamental limits in nonlocal and nonlinear nanophotonics, stimulating the generation of new plasmonic and optoelectronic applications.

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