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Plasmonic Enhanced Interaction of Light with Fluorophores and Graphene for Applications in Optics and Optoelectronics

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Abstract:

The main focus of this work is to investigate how surface plasmons can be used to enhance the interaction with light in two different physical systems: fluorophores, and graphene. Fluorophores are a powerful tool for the investigation of matter at a molecular or supramolecular level, in particular in the field of biology, as for instance by using fluorescent probes to image the structure and interplay of intracellular components. Within the scope of this thesis, the emission enhancement of fluorophores in the vicinity of dielectric coated thin metal films is investigated. The enhancement is found to be based on resonant energy transfer between fluorophores via weakly bound surface plasmon polaritons supported by the metal-dielectric structure. The accumulation of energy within a distribution of fluorophores in a small region above the coated metal film leads to the effect of optical sectioning comparable to what can be achieved using total internal reflection microscopy. The potential application of the findings is confirmed by a confocal microscopy study of cells grown on top of the metal-dielectric structures consisting of quartz plates coated with silver and silicon nitride. In addition to the sectioning effect, the coated substrates are used to perform axial fluorescence imaging with a resolution of down to 10 nm by measuring the variation in the emission spectrum of fluorophores with respect to their distance to the substrate. The potential of the imaging technique, termed color-coded optical nano-sectioning (COCOS), is demonstrated by imaging fixed cells, as well as by determining the axial position of fluorophores in living cells in a time-resolved manner.

Graphene is a single layer of carbon atoms arranged in a honeycomb lattice with exceptional electronic and optical properties. It is currently employed for the development of optoelectronic devices such as fast photodetectors in view of high-speed applications. Moreover, despite recent efforts, ultimate speed limitations of these detectors could not be determined. In the framework of this thesis, the intrinsic response time of metal-graphene photodetectors is measured using a nonlinear optical autocorrelation technique with ultrashort laser pulses. In contrast to a fully electronic approach, high-speed electronic equipment is not required. The response time, which is mainly given by the short lifetime of the photogenerated carriers, is found to be in the picosecond range corresponding to a bandwidth of hundreds of Gigahertz. Furthermore, the enhancement of graphene-light interaction using localized surface plasmons is investigated by analyzing the enhancement of Raman transitions in graphene. The plasmons are generated in silver nanoislands, which are fabricated by means of metal deposition directly on graphene. Despite the broad size distribution of the nanoislands, an almost 100-fold enhancement of the Raman signal can be obtained. An analytical model for the optical properties of the nanoislands, as well as for the enhancement factors of the Raman transitions is formulated and compared with experimental results. In addition, the doping in graphene introduced by the nanoislands is investigated, both optically and electrically.