## Theoretical Modeling of Nanoscale Confined Light: From Metal Nanoparticles to Nanoholes

Stephen K. Gray Chemistry Division and Center for Nanoscale Materials Argonne National Laboratory Argonne, Illinois 60439 Email: gray@tcg.anl.gov

Nanophotonics is about the manipulation of electromagnetic waves in nanoscale environments [1-3]. Often wavelengths greater than a few hundred nanometers are of interest. This implies that sub-diffraction limit interactions are needed in order for such light to be affected by nanoscale objects. Much work in nanophotonics attempts to utilize surface plasmons, i.e. electromagnetic surface waves consistent with collective electronic excitations near metal surfaces. Surface plasmons are evanescent, implying an effective imaginary component to the associated wave vectors, which can overcome the diffraction limit and lead to nanoscale confinement and/or propagation of electromagnetic energy. Metal nanoparticles (MNPs) have been frequently studied in relation to their surface plasmon properties. In particular, isolated MNPs can exhibit localized surface plasmons (LSPs), i.e., electromagnetic modes confined to the surfaces of the particles. Applications of LSPs to chemical and biological sensing have been suggested. Arrays of MNPs might also exhibit collective surface plasmons with traveling character, which could lead to nanoscale propagation of light suitable for optical wires. A more established way of generating traveling surface plasmons is to excite surface plasmon polaritons (SPPs) on thin metal films, and there is also much interest in such SPPs for nanophotonics applications.

In order to best understand the physical phenomena involved in nanophotonics, and to suggest new experimental directions, it is helpful to carry out realistic simulations using computational electrodynamics methods. The approach used here is the finite-difference time-domain (FDTD) method, which can be applied to a variety of complex situations [4]. LSPs and SPPs involve metallic regions of space described by dielectric constants with negative real parts. Naïve implementations of the FDTD method in such cases can be unstable or inaccurate. I show how to carry out realistic FDTD realistic simulations of surface plasmons and their properties such as optical cross sections [5]. The possible nanoscale localization of radiation in certain funnel-like arrays of silver nanowires is discussed [5], and also the role of dielectric coating [6]. All the calculations discussed so far involve exciting the MNPs with ordinary light. Total internal reflection (TIR) can also be used to excite MNPs. I discuss a joint experimental-theoretical study of TIR excitation of isolated MNPs and arrays of MNPs [7]. In particular the experiments obtain results consistent with enhanced low-angle scattering due to LSP resonances and the theoretical calculations are able to confirm that this is indeed the case.

As noted in the introductory paragraph, traveling surface plasmons or SPPs are also of great interest. It is now well established that nanoscale holes in metal nanofilms, when illuminated with ordinary light can generate SPPs [1]. I describe another joint experimental-theoretical study showing that the nanoholes act as point-like sources of SPPs in the plane of the metal surface and that a certain interference effect explains the experimentally observed intensity fringes [8]. A strength of the FDTD approach is that we could explicitly include the probe in some of our calculations and so we were also able to study how the probe affects the dynamics.

I conclude by indicating some new directions in this research program, including multiscale modeling and the coherent control of nanophotonics.

## Acknowledgment

This work was supported by the Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences, and Biosciences, U.S. Department of Energy, under Contract No. W-31-109-ENG-38.

## References

- [1] W. L. Barnes, A. Dereux and T. W. Ebbesen, *Nature* 424, 824 (2003).
- [2] S. A. Maier, P. G. Kik, H. A. Atwater, S. Meltzer, E. Harel, B. E. Koel, and A. A. G. Requicha *Nature Materials* **2**, 229 (2003).
- [3] A. V. Zayats and I. I. Smolyaninov, J. Opt. A: Pure Appl. Opt. 5, S16 (2003).
- [4] A. Taflove and S. C. Hagness, *Computational Electrodynamics: The Finite-Difference Time-Domain Method, 2nd ed.* (Artech House, Boston, 2000).
- [5] S. K. Gray and T. Kupka, *Phys. Rev. B* 68, 045415 (2003).
- [6] J. M. Oliva and S. K. Gray, Chem. Phys. Lett. **379**, 325 (2003).
- [7] G. A. Wurtz, J. S. Im, S. K. Gray, and G. P. Wiederrecht, *J. Phys. Chem. B* 107, 14191 (2003).
- [8] L. Yin, V. K. Vlasko-Vlasov, A. Rydh, J. Pearson, U. Welp, S.-H. Chang, S. K. Gray, G. C. Schatz, D. E. Brown, and C. W. Kimball, *Appl. Phys. Lett.* 85, 467 (2004).