Experiments in Spectroscopy

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Contents

Part I

Fundamentals

Part II

Two Level Systems

Part III

Nonlinear Spectroscopy

Part IV

Plasmonics

Chapter 17

Plasmonic Nano-Rods

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Tasks

- Model the angle-dependent extinction spectrum of an array of plasmonic particles, as measured by Simon Durst (Bayreuth) and shown below. Discuss the observed phenomena.
- Assume that the embedding medium contains a dye with a narrow Lorentzian absorption line. Model and discuss the dispersion relation.

How this is measured

Waveguides

Let us start by investigating the optical modes of a thin wire. We assume cylindrical symmetry and coordinates z and ρ. The wire has a radius a and is made of a material (first dielectric, later metal) of dielectric function ϵ_{in}. It is embedded in a dielectric matrix characterized by ϵ_{out}. We solve Maxwell's equation with these boundary conditions.[1]

The electric field is described inside and outside the wire by cylindrical Bessel J and Hankel $H^{(1)}$ functions of the first kind, respectively. The lowest order mode is[2]

$$\begin{aligned}\mathbf{E}_{in} &= E_0 \left(J_0(\kappa_1 r)\,\hat{\mathbf{z}} + \frac{ik_z}{\kappa_1} J_1(\kappa_1 r)\,\hat{\mathbf{r}} \right) e^{i(k_z z - \omega t)} \\ \mathbf{E}_{out} &= E_0 \left(H_0^{(1)}(\kappa_2 r)\,\hat{\mathbf{z}} - \frac{ik_z}{\kappa_2} H_1^{(1)}(\kappa_2 r)\,\hat{\mathbf{r}} \right) e^{i(k_z z - \omega t)}\end{aligned}$$

where k_z is the component of the wave bvector (length in vaccum $k_0 = 2\pi/\lambda$) along the wire. The components perpendicular to the wire are defined as

$$\kappa_i = k_0 \sqrt{\epsilon_i - (k_z/k_0)^2} \tag{17.1}$$

with $\epsilon_i = (\epsilon_{in}, \epsilon_{out})$. The boundary condition at $r = a$ leads to a condition for k_z (see also H/N eq. 12.53)

$$\frac{\epsilon_{in}}{\kappa_1 a} \frac{J_1(\kappa_1 a)}{J_0(\kappa_1 a)} - \frac{\epsilon_{out}}{\kappa_2 a} \frac{H_1^{(1)}(\kappa_2 a)}{H_0^{(1)}(\kappa_2 a)} = 0 \tag{17.2}$$

[1] Details see Saleh /Teich chapter fiber optics

[2] **Takahara**.

Guiding of waves requires that the electric field is localized near the wire, i.e., the radial component of the wave vector outside the wire κ_2 has to be imaginary, or $k_z/k_0 > \sqrt{\epsilon_{out}}$. At the same time, the wave should propagate inside the wire, i.e., k_z should be (almost[3]) real, or $\sqrt{\epsilon_{in}} > k_z/k_0$.

[3] When the dielectric functions are complex-valued, also k_z becomes complex-valued

For dielectric waveguides, the core has to have a higher index of refraction than the embedding medium. When the radius of the wire is decrease, the decay of the field outside the wire becomes slower and slower. By Fourier transformation from κ to ρ one finds a characteristic lower limit for radius R of the field distribution, independent of the wire diameter,

$$R > \frac{\lambda}{2\sqrt{\epsilon_{in}}} \tag{17.3}$$

Dielectric waveguides are this limited in their size of the mode field to approximately the wavelength in the core medium.

Plasmonic waveguides in contrast can become very small. When the dielectric function ϵ_{in} is negative, the mode field remains bound close to the wire even for small a wire radius. The downside is that losses increase. The wave vector in propagation direction becomes complex. The real part describes the effective index of refraction of the mode, the imaginary part the losses due to absorption in metal. These losses increase drastically when the wire becomes smaller, as shown in Fig XXX

Figure 17.1: Al wire alpha and beta similar to Fig. 12.20 in H/N

The large component of the wave vector in propagation direction k_z in a plasmonic waveguide corresponds to a short effective wavelength $\lambda_{in} = 2\pi/k_z$. The thinner the waveguide, the shorter the effective wavelength. Novotny shows that a linear relation to the vacuum wavelength exists XXX cite

$$\lambda_{in} = n_1 + n_2 \frac{\lambda}{\lambda_p} \tag{17.4}$$

where $\lambda_p = 2\pi c/\omega_p$ is the plasma wavelength of the Drude metal and the n_i are constants depending on geometry and refractive indices. [4]

[4] see eq. 14 in XXX

Side note: Leakage radiation

The component of the wave vector in propagation direction k_z is larger than the maximum possible length of a wave vector in the embedding medium $k_0\sqrt{\epsilon_{out}}$. This means that momentum conservation does not allow photons to leave the waveguide. Plane waves with such a value of k_z are evanescent (have an imaginary $k_\perp$) in the embedding medium. In this way, a mode can propagate without loses to the environment. However, this also hinders observation of such a propagation. One only could detect the emission at the end of the waveguide or at defects.

One way around is leakage radiation. When the waveguide is placed on or near a medium with a higher index of refraction $k_z < k_0\sqrt{\epsilon_{substrate}}$, then this substrate supports suitable free-space modes. The distance between waveguide and substrate defines the coupling (as seen by the observer) or the losses (from the point of view of the waveguide). In such an experiment one can see bright emission along the whole waveguide.

Fabry-Perot modes in nano-rods

We now cut out a piece of a thin plasmonic waveguide and call this object a nano-rod. The propagation of the plasmon mode along the waveguide is the same as in the preceding section, but now the wave is reflected back at the ends of the waveguide. The free space around the nano-rod does not support modes of sufficient high wave vector, so that the light can not just propagate out. The two ends thus form two mirrors of a Fabry-Perot cavity and the short piece of waveguide in between is similar to the medium in the cavity. We expect to find periodic resonances when varying the length of the rod. This is indeed what is observed. However, the apparent length of the rod is larger by some offset length L_o

$$L_{res} = n\,\frac{\lambda_{in}}{2} + L_o \tag{17.5}$$

The exact value of the offset L_o depends on details of the waveguide end, for example how this is rounded or cut flat, and also from where to where on measures the rod length L_{res}.

The physical origin of this apparent additional length L_o is that a propagating plasmon is a quasi particle combined of free electrons and an electromagnetic field. The electrons have to stay inside the metal to within less than a lattice constant. The optical field however extends all around the nano-rod and thus also extends over the ends of the rod. This gives an additional length.

In some publications this additional length L_o is called a reflection phase $\Delta\phi$, as one could also describe the process in terms of phases as

$$\frac{2L_{res}}{\lambda_{in}}\,2\pi = n\,2\pi + 2\Delta\phi \tag{17.6}$$

This is in analogy to the phase acquired by an optical beam undergoing total internal reflection. In this case, $k_{z,1}$ is real-valued, but $k_{z,2}$ is complex, as it describes an evanescent wave. The reflection coefficient

$$r^s_{12} = \frac{k_{z,1} - k_{z,2}}{k_{z,1} + k_{z,2}} \tag{17.7}$$

becomes then also complex, so that the field acquires a phase shift

$$\Delta\phi = \arg(r^s_{12}) \tag{17.8}$$

Field distribution inside and outside the nano-rod

Dipole model of the modes in a nano-rod

Far-field excitation of modes

Near-field excitation of modes

Nonlinear effects with modes

Appendices