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Coherent control of plasmons in nanoparticles with nonlocal response

D. McArthur, B. Hourahine, F. Papoff*

Department of Physics, SUPA, University of Strathclyde, 107 Rottenrow, Glasgow G4 0NG, UK

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ABSTRACT

We discuss a scheme for the coherent control of light and plasmons in nanoparticles that have nonlocal dielectric permittivity and contain nonlinear impurities or color centers. We consider particles which have a response to light that is strongly influenced by plasmons over a broad range of frequencies. Our coherent control method enables the reduction of absorption and/or suppression of scattering.

1. Introduction

Recent progress in nanophotonics and plasmonics has led to many important applications, including photovoltaics [1–3], sensors [4] and medicine [5]. Coherent control is a very topical subject in this area of research as it allows one to enhance the interaction of light with matter at the nanoscale: several groups have investigated nonlinear [6] and linear control based on pulse shaping [7–11], a combination of adaptive feedbacks and learning algorithms [12], as well as optimization of coupling through coherent absorption [13] and time reversal [14]. Coherent control of second-harmonic generation has been studied in nanowires [15,16] and nanospheres [17,18] while, in quantum optics, interference between fields was proposed as a way to suppress losses in a beam splitter [19] and has been recently applied to show control of light with light in linear plasmonic metamaterials [20]. These control methods have been applied only to systems with local responses. Particles with spatially nonlocal response behave very differently from particles with local response as they support irrotational charge density waves, such as plasmons, that do not radiate and can reach the central region of the particle over a large range of frequencies; on the contrary, particles with local responses support longitudinal modes only when the real part of the electric permittivity ϵ is null [21]. As a result, particles with nonlocal responses also exhibit a shift of the main resonance with respect to particles with local response for the same geometry and, in some metals, also have extra resonances at short wavelengths [22–27]. From the point of view of control, the main difference between media with local and nonlocal response is that in media with nonlocal response we can use light to control not only internal and scattered light, but also currents. In previous papers we have developed a coherent control theory for metallic nanospheres with diameters of at least 50 nm, for which nonlocal effects may be important only in a very thin layer at the boundary of the particles [17,18] where nonlinear processes take place. In this paper we investigate smaller nanoparticles in which the nonlinearity is due to an impurity, or color centers, inside the particle and for which nonlocal effects are important not only at the surface. We focus here on nanospheres because in this case the theory is fully analytical, but the approach we develop is based on the interference of fields at the surface of the particle and can be applied whenever longitudinal and transverse waves are both allowed, independently of the shape of the particle or the origin of the longitudinal waves. In particular, systems such as core-shell spherical particles, with diameters of 50 – 100 nm and an external layer with nonlocal response of a few nanometers, have similar properties to the spheres we consider here and interact more strongly with light. Consequently, these types of systems would be better from the point of view of applications. In this case, the control can be modeled similarly to the control method employed here by using the Mie theory for layered spheres [28,29]. However, depending on the materials used, there could be electron spill-out between the inner core and outer layer which would need to be included in the modeling of nonlocality [30].

We develop coherent control techniques which are extremely sensitive to phase variations and produce a reduction of the

* Corresponding author.
E-mail addresses: duncan.mcarthur@strath.ac.uk (D. McArthur), f.papoff@strath.ac.uk (F. Papoff).

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absorption and variations of the scattered energy or of the amplitudes of the plasmons over several orders of magnitude. These unusual features enable applications such as detection of deeply sub wavelength changes in the position of the particle, reduction of dissipation, suppression of radiative losses, sensing of variations in the electric permittivity ϵ and magnetic permeability μ and optical routing.

2. Including nonlocality in Maxwell's equations

When one of the characteristic dimensions of the particle/structure is of the order of the electron free path, the free current is governed by a nonlocal equation that admits longitudinal waves. In the hydrodynamical model [22–27], the nonlocal response is modeled semi-classically by considering the free charges in the metal as a fluid governed by the linearized Navier–Stokes equation and with a pressure term that has a quantum origin and is proportional to the Fermi velocity. The interaction of the particle with light is then given by Maxwell's equations combined with the linearized Navier–Stokes equations [31],

$$\nabla \times \boldsymbol{E} = -\mu \partial_t \boldsymbol{H}, \tag{1}$$

$$\nabla \times \boldsymbol{H} = \partial_t \left[\epsilon_b \boldsymbol{E} + \boldsymbol{P}_f \right], \tag{2}$$

$$\left(\partial_{tt} + \gamma_f \partial_t - \beta^2 \nabla \nabla \cdot \right) \boldsymbol{P}_f = \epsilon_0 \omega_p^2 \boldsymbol{E}, \tag{3}$$

where $\boldsymbol{E}$, $\boldsymbol{H}$ are the electric and magnetic fields, $\boldsymbol{P}_f$ is the polarization due to the free current density $\boldsymbol{J}_f$ $(\partial_t \boldsymbol{P}_f = \boldsymbol{J}_f)$, ϵ_b is the electric permittivity due to the bound charges, γ_f is the damping factor due to collisions of the free charges, ω_p is the plasma frequency of the material and $\beta^2 = (3/5)v_F^2$, with v_F the Fermi velocity. The nonlocal term $\nabla\nabla \cdot \boldsymbol{P}_f$ in (3) affects the interaction of the particle with light in two ways. First, the electric field contains both a transversal part $\boldsymbol{E}_T (\nabla \cdot \boldsymbol{E}_T = 0)$ and a longitudinal part $\boldsymbol{E}_L (\nabla \times \boldsymbol{E}_L = 0)$, each with it's own dispersion relation. The longitudinal waves are expanded in terms of the longitudinal solutions of the Helmholtz equation and are associated with charge density waves, such as plasmons, but not to radiation as $\boldsymbol{E}_L$ is decoupled from time-dependent magnetic fields. Secondly, (3) also modifies the interaction with light through an additional boundary condition that is necessary to determine $\boldsymbol{P}_f$. This boundary condition is considered together with the usual continuity of the tangent components of $\boldsymbol{E}$ and $\boldsymbol{H}$ [32]. In media that do not support a surface density of free charges, the component of the free current density normal to the boundary of the particle is continuous [26,27]. At a dielectric-metal interface, this condition implies that the normal component of $\boldsymbol{P}_f$ in the metal has to vanish at the boundary, as dielectrics do not support free currents. Using the integral version of the divergence and an infinitesimal pillbox on the right-hand side of (2) shows that the normal component of $\epsilon_b \boldsymbol{E} + \boldsymbol{P}_f$ is also continuous at the boundary. Therefore the continuity of the normal component of $\boldsymbol{P}_f$ is equivalent to the continuity of the normal component of $\epsilon_b \boldsymbol{E}$, which provides the additional boundary condition,

$$\hat{\boldsymbol{n}} \cdot \epsilon_b^i \boldsymbol{E}^i = \hat{\boldsymbol{n}} \cdot \epsilon_b^e \left(\boldsymbol{E}^s + \boldsymbol{E}^0 \right), \tag{4}$$

where ϵ_b^e, ϵ_b^i are the permittivity due to bound charges of the external and internal media respectively, and $\boldsymbol{E}^i$, $\boldsymbol{E}^s$, $\boldsymbol{E}^0$ are the internal, scattering and incident fields. From (2) we have $\nabla \cdot \boldsymbol{P}_f = -\nabla \cdot \epsilon_b \boldsymbol{E}$, which can be used to find the dispersion relation of the longitudinal waves and to recast the Maxwell equations in terms of the longitudinal and transverse electric fields; we then use the boundary conditions to determine the amplitudes of the longitudinal and transverse waves. The additional boundary condition in (4) and the presence of $\boldsymbol{E}_L$ lead to modified Mie coefficients [33] for the sphere.[1]

3. Mode structure

At the heart of our theory is the Stratton-Chu representation theorem that allows one to express any internal and scattered fields of any smooth (possibly inhomogeneous) particle in terms of integral operators acting on the electromagnetic fields at the surface of the particles [34]. In practice this means that the response of a particle to light generated by impressed driving sources (which are either internal or external) can be determined by expanding the internal and the scattered fields in terms solutions of Maxwell's equations for the internal and the external media that can approximate any field incident to the surface of the particle from the inside or the outside with arbitrary precision [35]. By defining surface fields with the electric and magnetic component parallel to the surface of the particle as $\boldsymbol{f} \equiv \left[-\hat{\boldsymbol{n}} \times \left(\hat{\boldsymbol{n}} \times \boldsymbol{E} \right), -\hat{\boldsymbol{n}} \times \left(\hat{\boldsymbol{n}} \times \boldsymbol{H} \right) \right]^T$, where T means transpose, $\hat{\boldsymbol{n}}$ is the unit vector normal to the surface, and the scalar product of two surface fields is the overlap integral $\boldsymbol{f}_1 \cdot \boldsymbol{f}_2 = \sum_{i=1}^3 \int_S \boldsymbol{f}_{1_i} \boldsymbol{f}_{2_i}^* ds$, where the index i labels the components of an arbitrary system of coordinates, the coefficients of the Mie modes of a sphere with local response are determined by projecting the incident fields on these modes using analytical formulae based on the scalar product defined on the surface [36]. These formulae apply also to particles whose modes can be found only numerically [35] and are very useful to determine the phase and amplitude of coherent light sources in order to modify the linear [36] and nonlinear [17,18] response of nanoparticles.

For particles with nonlocal response, one has also to include a complete set of longitudinal modes of the electric field corresponding to the plasmons; the coefficients for transverse and longitudinal modes can be calculated by fulfilling the continuity of the transverse component of the electric and magnetic fields as well as the boundary condition in (4) on the normal component of the electric field. To take into account (4), we need to also include the normal part of the electric field in the definition of the surface fields so that they now have five components,

$$\boldsymbol{f} \equiv \left[\hat{\boldsymbol{n}} \cdot \epsilon_b^{i/e} \boldsymbol{E}, -\hat{\boldsymbol{n}} \times \left(\hat{\boldsymbol{n}} \times \boldsymbol{E} \right), -\hat{\boldsymbol{n}} \times \left(\hat{\boldsymbol{n}} \times \boldsymbol{H} \right) \right]^T, \tag{5}$$

where ${\epsilon_b}^e$ is used for scattered and external fields, and ${\epsilon_b}^i$ is used for the internal field.[2] The scalar product between surface fields is modified accordingly, and is given by the sum of the overlap integrals of these five components. As a consequence of the spherical symmetry, only modes with the same value of l (orbital angular momentum) and m (angular momentum along z) can couple. For each value of l and m, using the angular dependence, one can group the modes into two sets: the set of transverse electric modes—as in a sphere with local response—and a set of two internal modes (the transverse magnetic and the longitudinal mode) and the transverse magnetic scattering mode. In a sphere the analysis can be limited to sets of modes with the same l and m because two modes with different l or m are orthogonal. For non

[1] Note that in [22] the authors claim that it is not physically possible to distinguish between free and bound charges and, therefore, that it should be the normal component of the total current which is continuous at the surface. Using a similar approach as the one above, this assumption is equivalent to the continuity of the normal component of the electric field [23]. There is very limited difference in the numerical results given by these two additional boundary conditions and no experimental evidence to support one over the other.

[2] Using the additional boundary condition described in [22,23], the first component of (5) should be replaced by $\hat{\boldsymbol{n}} \cdot \boldsymbol{E}$