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Peculiarities of light scattering by nanoparticles and nanowires near plasmon resonance frequencies

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Abstract. Results related to the peculiarities of light scattering by nanoparticles and nanowires near plasmon resonance frequencies are reported. It is shown that the scattering problem for weakly dissipative media cannot be analyzed in the dipole approximation, in contrast to the classical Rayleigh scattering. A structure of the Poynting vector in the near field area is obtained. It is shown that small variations of the size parameter or/and the incident light frequency may lead to drastic transformations of the near field distribution.

1. Introduction

Modern nanotechnology and nanooptics deal with objects whose size is smaller (or even much smaller) than the radiation wavelength λ . Thus, the actual problem for nanotechnology, data storage technology and nanopatterning with laser ablation is related to the ability to overcome the diffraction limit. Many ideas have been suggested, including SNOM technique and other techniques using laser in combination with a tip of AFM, etc. [1-4]. Lately much attention has been paid to the methods of plasmonics [5], related to excitation of surface electromagnetic waves within different nanostructures. These structures are used in many modern applications, e.g. in super-RENS [6], heat assisted magnetic recording (HAMR) [7], etc.

In the present paper we discuss the peculiarities of light scattering by nanoparticles and nanowires near plasmon resonance (or polariton resonance in dielectrics) frequencies. We will show that in the specified regions light scattering undergoes dramatic changes relative to the conventional Rayleigh scattering and a wide variety of new effects comes into being, provided the nanoobjects consist of weakly dissipating media. For such media the radiative damping [8] related to inverse transformation of localized plasmons generated by incident light into scattered electromagnetic field plays an important role and for this reason the scattering problem cannot be treated in the dipole approximation. It was demonstrated for spherical particles in Refs. [8-11]. A similar effect for nanowires is discussed in the present paper. The extinction coefficient for a spherical particle may demonstrate the inverse hierarchy of optical resonances [8, 10, 11], so that the resonant extinction cross-section increases with an increase of the resonance order (dipole, quadrupole, etc.) in contrast to its dramatic decrease for higher order resonances in the case of the Rayleigh scattering. We name this effect the anomalous scattering [10, 11]. The anomalous scattering results in the unusual and complicated structure of the electromagnetic field in the vicinity of the particle including optical vortexes and other peculiarities [9].

2. **Anomalous light scattering by a spherical particle**

Though light scattering by a spherical particle is one of the most fundamental problems of classical electrodynamics, the general physical understanding of the problem has not changed much since the publication of its exact solution by Mie in 1908 [12]. As for light scattering by a particle whose size is much smaller than the wavelength of incident light, its understanding up to now is based upon the approach developed by Lord Rayleigh in 1871 [13]. According to the approach a small particle should emit electromagnetic radiation as an oscillating electric dipole (the Rayleigh scattering). The point to be made is that this simple description has quite a general and very important exception, when the scattering process has very little in common with the Rayleigh scattering, and the extinction (scattering) cross-section differs from that given by the Rayleigh approximation in orders of magnitude.

Let us elucidate this exception. The simple formula for the Rayleigh approximation can be easily found from the general Mie solution. According to this solution, the extinction, scattering and absorption cross-sections are given by the following expressions [14]:

$$\sigma_{ext} = \frac{2\pi}{k_m^2}\sum_{\ell=1}^{\infty}(2\ell+1)\mathrm{Re}(a_\ell + b_\ell),\ \sigma_{sca} = \frac{2\pi}{k_m^2}\sum_{\ell=1}^{\infty}(2\ell+1)\left\{|a_\ell|^2 + |b_\ell|^2\right\},\ \ \sigma_{abs} = \sigma_{ext} - \sigma_{sca}. \qquad (1)$$

Here we consider a spherical particle with complex refractive index n_p and radius a placed in unbounded medium with purely real refractive index n_m, and $k_{m,p} = 2\pi\, n_{m,p}/\lambda$ are the corresponding wave numbers. The scattering electric a_ℓ and magnetic b_ℓ amplitudes are defined by the Mie formulae [14]

$$a_\ell = \frac{n\psi_\ell'(q)\psi_\ell(nq) - \psi_\ell(q)\psi_\ell'(nq)}{n\varsigma_\ell'(q)\psi_\ell(nq) - \psi_\ell'(nq)\varsigma_\ell(q)}\ ,\quad b_\ell = \frac{n\psi_\ell'(nq)\psi_\ell(q) - \psi_\ell(nq)\psi_\ell'(q)}{n\psi_\ell'(nq)\varsigma_\ell(q) - \psi_\ell(nq)\varsigma_\ell'(q)}, \qquad (2)$$

where $q = 2\pi\, a\, n_m/\lambda$, $n = n_p/n_m$, $\psi_\ell(z) = (\pi z/2)^{1/2} J_{\ell+1/2}(z)$, $\varsigma_\ell(z) = (\pi z/2)^{1/2} H^{(1)}_{\ell+1/2}(z)$, $J_\nu(z)$ and $H_\nu^{(1)}(z) = J_\nu(z) + iN_\nu(z)$ are the Bessel and the Hankel functions. The prime denotes derivative over the entire function's argument, i.e. $\psi_\ell'(z) \equiv d\psi_\ell(z)/dz$, etc.

Using expansions of these special functions in power series for a small particle, where $q << 1$ one arrives at the classical Rayleigh formula [14]:

$$\sigma_{sca} = \frac{8}{3}\pi a^2 q^4 \left|\frac{\varepsilon - 1}{\varepsilon + 2}\right|^2, \qquad (3)$$

where $\varepsilon = n_p^2/n_m^2$ stands for relative dielectric permittivity.

Expression (3) has a resonant denominator, which diverges at $\varepsilon = -2$. The physical meaning of the divergence is well known and quite clear. The particle could be regarded as a resonator. It has its own eigenmodes (surface localized plasmons). At $\varepsilon(\omega) = -2$ the frequency ω of the dipole eigenmode equals the one for the incident light. Thus, we face nothing but trivial resonance. If the dissipation rate is zero there is no damping and the amplitude of the resonant mode should diverge. The conventional way to avoid the divergence is to include finite dissipation. Since any actual physical system must have some dissipative mechanisms $\mathrm{Im}\,\varepsilon(\omega)\neq 0$ and expression (2) remain finite at any ω.

However, in addition to the dissipative damping the problem in question must possess another mechanism existing even at zero dissipation rates [8]. Namely, the temporal oscillations of the field related to excitation of eigenmodes result in oscillations of the corresponding polarizations (dipole, quadrupole, etc.) of the particle, i.e. to emission of electromagnetic waves, which transfer energy from the particle to infinity. It means that particle eigenmodes always are *damped*. Being entirely related to inverse transformation of localized plasmons into propagating electromagnetic radiation the radiative damping has nothing to do with dissipation and it exists even at purely real $\varepsilon_p(\omega)$. The radiative damping means the eigenfrequencies always have non-zero imaginary part, i.e. the exact resonance cannot occur at any purely real frequency ω. In other words, it means that at small enough dissipation rates equation (3) becomes erroneous at any (as small as desired) radius of the particle. In this case formula (3) should be replaced by another expression, obtained from the exact Mie solution of the problem which includes the radiative damping explicitly. The parameter region where the radiative damping prevails over the dissipative is exactly the one where the anomalous scattering is realized. Note in this connection recent publication [15], where the applicability conditions of the dipole approximation for light scattering by small particles are also discussed and inequalities similar to those of Ref. [8] are obtained.

Analysis shows that in Eq. (1) only amplitudes a_ℓ have resonant denominators. These amplitudes can be presented as $a_\ell = \Re_\ell/(\Re_\ell + i\Im_\ell)$ where $\Re_\ell$ is used for the numerator of a_ℓ in Eq. (2). When the dielectric permittivity $\varepsilon = n^2$ is a real quantity (i.e. $\mathrm{Im}\,\varepsilon = 0$) functions $\Re_\ell$ and $\Im_\ell$ are also purely real. Far from the plasmon resonance $|\Im_\ell| >> |\Re_\ell|$, thus $a_\ell \approx -i\Re_\ell/\Im_\ell$, which at small q leads to the Rayleigh formula (3). However the exact plasmon resonance corresponds to the situation when $\Im_\ell(n,q)=0$. For small q this equation can be presented in the following form [11]

$$n^2 + \frac{\ell+1}{\ell} - \frac{q^2}{2}\left(n^2-1\right)\left[\frac{n^2}{2\ell+3} + \frac{\ell+1}{\ell(2\ell-1)}\right] + \ldots = 0\,, \tag{4}$$

Note that $\varepsilon = n^2 = n^2(\omega)$, thus, Eq. (4) determines the resonant frequencies $\omega = \omega_\ell$ for the particle with small but finite size; these frequencies can be found from Fig. 1. Owing to vanishing of $\Im_\ell$ it is clear that correct description of the resonances requires accounting of $\Re_\ell \propto q^{2\ell+1}$ in the resonant denominators. This term is responsible for the shift of the poles of the scattering amplitude from the axis of real ω to the complex plane (radiative damping), which results in finite and real $a_\ell = 1$ at the resonant frequencies. As a result we obtain a simple expression

$$\sigma_{ext} = \sigma_{sca} \approx (2\ell+1)\frac{2\pi}{k_m^2}\,. \tag{5}$$

In contrast to the usual Rayleigh scattering, where $\sigma_{sca} \propto \omega^4$, Eq. (5) gives rise to the dependence $\sigma_{sca} \propto \omega^{-2}$. According to the Rayleigh approximation, the basic scattering is related to the dipole mode: all higher order resonances (quadrupole, etc.) are suppressed dramatically. In contrast to that, according to Eq. (5) anomalous scattering has the inverse hierarchy – the greater the order of resonance, the greater the corresponding cross-sections [8, 10, 11]. When we have dissipative media with $\mathrm{Im}\,\varepsilon_p \neq 0$ the resonance denominators in expression for a_ℓ have two types of imaginary terms, one related to the usual dissipation and the other related to the radiative damping. Then we immediately see that there is competition between the discussed radiative damping and the usual dissipative one. The discussed effects are observable provided the radiative damping prevails over the dissipative one. It results in the following applicability condition:

$$\varepsilon''(\omega) << \frac{q_m^{2\ell+1}}{\ell[(2\ell-1)!!]^2} . \qquad (6)$$

The right hand side of Eq. (6) decreases sharply with a decrease of q_m. It puts a certain constraint on the minimal size of the particle, when the anomalous scattering can be observed. At any as small as desired but finite $\varepsilon''(\omega)$ there is a certain quantity $\ell_{Ra}(\varepsilon'')$ so that at $\ell > \ell_{Ra}(\varepsilon'')$ Eq. (5) is violated, the anomalous scattering is suppressed and the normal Rayleigh hierarchy of the resonances is restored.

An example of inverse hierarchy of optical resonances for an aluminium particle as well as restoration of the normal Rayleigh hierarchy at large ℓ is shown in Fig. 2. For these calculations we used optical constants for the bulk material from [16] and took into account the variation of collision frequency with particle size: $\gamma_{eff}(\omega) = \gamma(\omega) + v_F/a$ [17]. We used $v_F = 10^8$ cm/s, which corresponds to the mean value of the anisotropic Fermi velocity in aluminum [18]. The inverse hierarchy of resonances as well as crossover to the normal Rayleigh hierarchy at large enough ℓ are seen clearly.

Though the inverse hierarchy of resonances is quite an appealing phenomenon, the most exciting feature of the anomalous scattering is absolutely unusual near-field distribution in the vicinity of the resonances. Regarding previous publications related to the problem in question, we may refer to paper [19], where the near-field of vector Poynting is calculated in the Rayleigh dipole approximation.

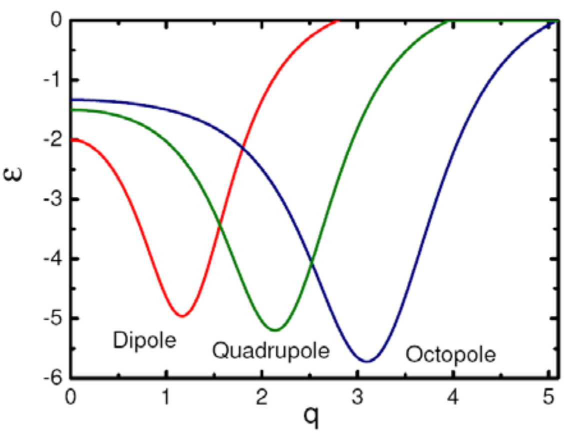

Fig. 1. Trajectories of the first three resonances with $\ell = 1$ (dipole), $\ell = 2$ (quadrupole) and $\ell = 3$ (octopole) versus size parameter q. At $q \to 0$ these resonances tend to $\varepsilon = \varepsilon_\ell = -(\ell+1)/\ell$. At small but finite q curves follow Eq. (4) and deviate to $\varepsilon < \varepsilon_\ell$. At $q \approx$ 1.167, $\varepsilon \approx 4.96$ for dipole (and similar to other resonances) the anomalous scattering resonance is merged with the nearest Mie resonance. Then resonant ε increases with a further increase in q beyond $q >$ 1.167.

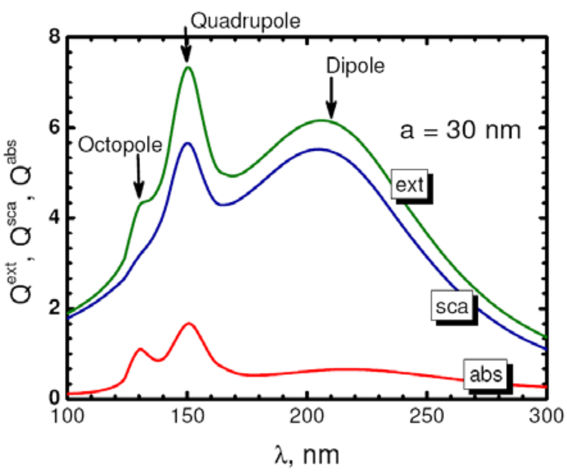

Fig. 2. Inverse hierarchy of resonances for an aluminum particle in vacuum. Normalized extinction, scattering and absorption cross-sections, $Q^i = \sigma^i / \pi a^2$, for a particle with $a =$ 30 nm as functions of incident light wavelength λ. Points of dipole, quadrupole and octopole resonances are indicated with arrows. The extinction cross-section of the quadrupole resonance is the greatest. With $a = 40$ nm the octopole resonance becomes the greatest [11].

The near-field distribution for the anomalous scattering differs drastically from that following for the same particle and the same order of resonance in the Rayleigh approximation [9]. The near-field has rather a complicated structure, which changes a lot with fine changes of the problem parameters, especially with changes of ω. A typical picture of the near-field with optical vortices is shown in Fig. 3. Unfortunately for lack of space we cannot discuss here this interesting matter in detail. Such a discussion will be presented in our forthcoming publications.

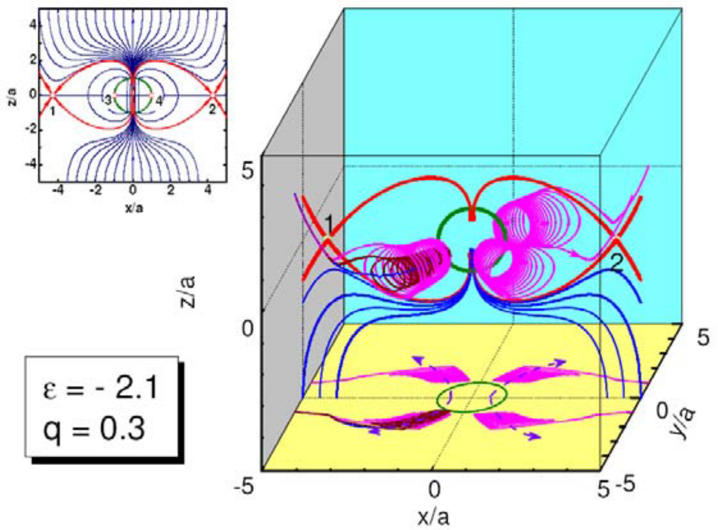

Fig. 3. Poynting vector lines for non-dissipative case around the spherical particle (incident plane wave with $\mathbf{E}\|x$ comes from $z=-\infty$). Left insert shows 2D field in *xz* plane. Points 1 and 2 are saddles. Thick red lines indicate the separatrixes in *xz*-plane. Field lines in 2D picture demonstrate circular energy flows around centers (points 3 and 4). In 3D plot one can see the energy flow outward the particle (helicoidally shaped fields lines). It illustrates the radiative losses of energy, general directions of which are shown by arrows on the bottom *xy* projection plane.

3. Anomalous light scattering by nanowires

Effects related to radiative damping are important also for nanowires with surface plasmons. This also leads to deviation of extinction and scattering coefficients from approximations for linear dipole, e.g.

$$\sigma_{sca} = 2aL\frac{\pi^2}{4}\left(\frac{\varepsilon-1}{\varepsilon+1}\right)^2 q^3, \tag{7}$$

where factor $2aL$ presents the geometrical cross-section, $L >> a$ is the length of the cylinder. Corresponding resonances arise at $\operatorname{Re}\varepsilon = -1$, where σ_{sca} divergent at $\operatorname{Im}\varepsilon \to 0$. However, owing to the radiative damping the divergence at $\operatorname{Im}\varepsilon = 0$ is cuts off and we arrive to formula, similar to Eq. (5): $\sigma_{sca} = 16L/k_{res}$, where k_{res} presents the wave vector at the resonant frequency. Compared to a spherical particle the near-field structure for nanowire can be more complicated see e.g. Fig. 4.

In both the cases (the spherical particle and the nanowire) the near-field structure turns out to be quite sensitive to fine detuning of frequency of the incident light from the exact resonant frequencies [20, 21]. Numerous applications of the anomalous scattering in nanotechnologies and related fields may be associated with (i) an enormous amplification of the incident electromagnetic field in the near-field area whose size is much smaller than the incident radiation wavelength; (ii) controllable changes of the near-field structure with changes of the incident light frequency; (iii) comparable intensity of the resonant electromagnetic field at different resonant frequencies of the incident light, corresponding to different orders of resonance, accompanied by quite a different field distribution for each order of the resonance. This is the proper way for optical manipulation in the field structure in the nanoscale region, which can be used for different applications in nanooptics.

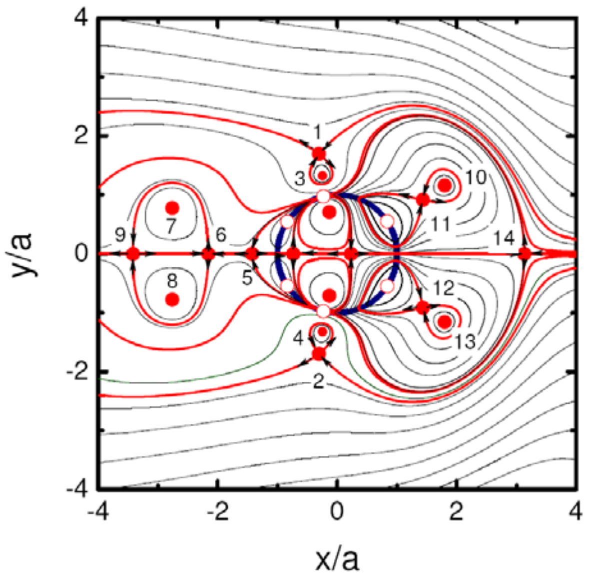

Fig. 4. Poynting vector field lines around the nanowire (cross-section in *xy*-plane) with $q = 1$ and $\varepsilon = -1$ (incident plane wave with $\mathbf{E} \| \mathbf{y}$, comes from $x = \infty$). Separatrixes are indicated by red lines. Enumerated 14 singular points around the wire correspond to zero values of Poynting vector components; points 1, 2, 5, 6, 9, 11, 12 and 14 are saddles, while points 3, 4, 7, 8, 10 and 13 are centers. There are also six singular points (open circles) on the wire surface: four centers (circular energy flow around these centers produce plasmon emission, responsible for radiative damping) and two saddles. Four singular points also exist inside the wire: two centers and two saddles.

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