Absorbing and emitting optical properties of a spherical plasmonic nanoantenna are described in terms of the size dependent resonance frequencies and damping rates of the multipolar surface plasmons (SP). We provide the plasmon size characteristics for gold and silver spherical particles up to the large size retardation regime where the plasmon radiative damping is significant. We underline the role of the radiation damping in comparison with the energy dissipation damping in formation of receiving and transmitting properties of a plasmonic particle. The size dependence of both: the multipolar SP resonance frequencies and corresponding damping rates can be a convenient tool in tailoring the characteristics of plasmonic nanoantennas for given application. Such characteristics enable to control an operation frequency of a plasmonic nanoantenna and to change the operation range from the spectrally broad to spectrally narrow and vice versa. It is also possible to switch between particle receiving (enhanced absorption) and emitting (enhanced scattering) abilities. Changing the polarization geometry of observation it is possible to effectively separate the dipole and the quadrupole plasmon radiation from all the non-plasmonic contributions to the scattered light.

Keywords: surface plasmon (SP) resonance, plasmon damping rates, multipolar plasmon modes, Mie theory, optical properties of gold and silver nanospheres, noble metal nanoparticles, receiving and emitting nanoantennas, dispersion relation, nanophotonics, SERS technique.
tromagnetic approach based on the analysis of the SP dispersion relations [28,34,36,37]. We treat the possibility of excitation of SP oscillations and their damping as a basic, intrinsic property of a conducting sphere. We provide some ready-to-use surface multipolar plasmon size characteristics of the dipole and higher polarity plasmon resonances in gold and silver nanospheres up to the radius of 150 nm covering the multipolar plasmon resonance frequencies in the range of about 1–4 eV for silver and 1.2–2.7 eV for gold nanoparticles. Optical properties of these metals are described by realistic, frequency dependent refractive indices of corresponding pure metals [38]. The multipolar SPs resonance frequencies and the SPs damping rates are treated consistently.

Implications of damping processes in SP applications are extremely important [4,39,40]. In this paper, we underline the role of the plasmon radiation damping in comparison with the internal energy dissipation damping in formation of receiving and transmitting properties of a plasmonic particle. In particular, we demonstrate that the enhancement factor of absorbing and scattering particle abilities depends on the relative contribution of the energy dissipative and radiative processes to the total damping of SP oscillations. These processes are size dependent.

We show that only knowing the size dependence of both: the multipolar SP resonance frequency and corresponding damping rate, an effective description of plasmonic properties is possible. SP size characteristics we study here allow not only to predict a frequency of SP resonance in given polarity order, but also to exploit a plasmonic particle as an effective receiving (enhanced absorption) or emitting (enhanced scattering) nanoantenna. These features are crucial for SPs applications.

2. Modelling SPs inherent size characteristics

Collective motion of surface free-electrons in a metallic particle can be excited by the EM field under the resonance conditions which is defined by intrinsic properties of a plasmonic sphere due to its size, optical (conductive) properties of a metal and dielectric properties of the sphere’s environment. The SP resonance takes place when the frequency of the incoming light \( \omega \) approaches at least one of the characteristic eigenfrequencies \( \omega_l (R), l = 1,2,3,... \) of a sphere of the radius \( R \). The dynamic plasmon charge density distribution, induced by EM field, can be quite complicated. With the increases of the sphere surface, the surface charge density distributions of polarity higher than the dipole one \( (l = 1) \) come into play [34].

The collectively oscillating electrons of SPs at curved surface must emit EM energy through radiation. Radiative damping is, then, the inherent property of SPs. It enables the enhancement of the EM field scattered by a sphere and is inseparably associated with SP oscillations at characteristic frequencies of multipolar plasmon modes.

To find the SP intrinsic size characteristics we use a self-consistent rigorous EM approach based on Ref. 36, and described in more details in Refs. 28, 29, and 34. We consider continuity relations at the spherical boundary for the tangent component of the transverse magnetic (TM) solution of the Helmholtz equation in spherical coordinates, while TM solutions only possess nonzero normal to the surface component of the electric field \( E_z \). This component is able to couple with the charge densities at the boundary. Resulting conditions define the dispersion relations in spherical coordinates [28,34]

\[
\sqrt{E_{in}(\omega) \xi_l (k_{out}(\omega) R)} \psi_l (k_{in}(\omega) R) - \sqrt{E_{out}(\omega) \xi_l (k_{out}(\omega) R)} \psi_l (k_{in}(\omega) R) = 0
\]

which are fulfilled for the complex eigenfrequencies of the fields \( \Omega_l, l = 1, 2, 3,... \) at the \( r = R \) distance from a sphere centre. \( \psi_l (z) \) and \( \xi_l (z) \) are Riccati-Bessel spherical functions, the prime marker (’') indicates differentiation with respect to the function’s argument, \( k_{in} = \omega/c \sqrt{E_{in}(\omega)} \) and \( k_{out} = \omega/c \sqrt{E_{out}} \).

The realistic modelling of the SP characteristics is possible if the functional dependences of the dielectric function on frequency in analytical form for both the sphere \( \varepsilon_{SP}(\omega) \) and the surrounding medium \( \varepsilon_{medium}(\omega) \) are known. It assures the correct coupling of the metal dispersion to the overall frequency dependence of the plasmon dispersion relation of Eq. (1). We used the modified dielectric function of the Drude electron gas model based on the kinetic gas theory \( \varepsilon_{in}(\omega) = \varepsilon_D(\omega) = \varepsilon_{\infty} - \omega_p^2 / \left( \omega^2 + i \gamma \omega \right) \) which quite well describes the optical properties of many metals within relatively wide frequency range [38]. \( \varepsilon_{\infty} \) is the phenomenological parameter describing the contribution of the bound electrons to the polarizability [41] which equals to 1 only if the conduction band electrons are responsible for the optical properties of a metal (e.g. sodium) [28]. For gold and silver, the interband transitions are important for defining their optical properties. \( \omega_p \) is the bulk plasmon frequency, \( \gamma \) is the phenomenological relaxation constant of the bulk material. For a perfect free-electron bulk metal with infinite boundaries, electron relaxation is due to electron-electron, electron-phonon, and electron-defect (grain boundaries, impurities, and dislocations) scattering processes. \( \gamma \) results from the average of the respective collision frequencies of electrons and thus, it is closely related to the electrical resistivity of the metal [24]. The functions \( \varepsilon_{in}(\omega) \) with the effective parameters \( \varepsilon_{\infty} = 9.84 \), \( \omega_p = 9.096 \text{ eV} \), \( \gamma = 0.072 \text{ eV for gold} \), and \( \varepsilon_{\infty} = 37 \), \( \omega_p = 8.9 \text{ eV} \), \( \gamma = 0.021 \text{ eV for silver} \) satisfactorily reproduce the experimental values of \( \text{Re}[\varepsilon_n(\omega)] \) [\( \text{Im}[\varepsilon_n(\omega)] \)] in the frequency ranges 0.8–5.0 eV for gold and 0.8–4.2 (4.0) eV for silver (Fig. 1). As illustrated in Fig. 1, the agreement of the model and measured frequency dependence of the dielectric function is very good, with exception of \( \text{Im}[\varepsilon_{in}(\omega)] \) dependence for silver at higher frequencies of the studied frequency range [above 4 eV, it is for some wavelengths outside the visible range \( \lambda < 31 \text{ nm} \)]

In the calculations, the optical properties of the dielectric medium outside the sphere are described by \( \varepsilon_{out} = n_{out}^2 \) with \( n_{out} \) chosen here to be 1 or 1.5. However, the formal-
Fig. 1. Real and imaginary parts of the dielectric function with effective parameters $\varepsilon_{\text{in}} = 9.84$ eV, $\omega_p = 9.096$ eV, $\gamma = 0.072$ eV for gold, and $\varepsilon_{\text{in}} = 3.7$ eV, $\omega_p = 8.9$ eV, $\gamma = 0.021$ eV for silver. Squares and circles are the corresponding values of $Re(n^2)$ and of $Im(n^2)$ (after Ref. 38).

The extreme values of $\omega^l_\ell(R)$ and $\omega^p_\ell(R)$ can be found from an approximated and very rough consideration using the power series expansion of the spherical Bassel and Hankel functions

$$j_l(z) = \frac{z^l}{(2l+1)!!} \left[ 1 - \frac{0.5z^2}{1!(2l+3)} + \frac{(0.5z^2)^2}{2!(2l+3)(2l+5)} - \cdots \right] \quad (2)$$

$$h_l(z) = -i \frac{(2l-1)!!}{z^{l+1}} \left[ 1 - \frac{0.5z^2}{1!(1-2l)} + \frac{(0.5z^2)^2}{2!(1-2l)(3-2l)} - \cdots \right], \quad (3)$$

where $(2l \pm 1)!! = 1 \times 3 \times 5 \times \cdots \times (2l \pm 1)$. Applying the limit of small arguments $z$ (so-called “quasistatic approximation”), and keeping only the first terms of the power series Eqs. (2) and (3) (what is not justified for the larger $l$ values), the dispersion relation of Eq. (1) is fulfilled under the condition below

$$-\frac{l}{l+1} \frac{\varepsilon_{\text{in}}}{\varepsilon_{\text{out}}} = 1. \quad (4)$$

For equation $\varepsilon_{\text{out}}(\omega)$ in the form given by Eq. (1), one gets

$$\omega^l_\ell = \left\{ \frac{\omega_p^2}{\varepsilon_{\text{in}} + [l(l+1)]\varepsilon_{\text{out}}} \right\}^{1/2}, \quad (5)$$

$$\omega^l_\ell = -\frac{\gamma}{2}. \quad (6)$$

Neglecting relaxation ($\gamma = 0$) for a perfect free-electron metal ($\varepsilon_{\text{in}} = 1$) and $\varepsilon_{\text{out}} = 1$, Eq. (5) leads to the well-known multipolar plasmon frequency values of a metal sphere within the “quasistatic approximation” [37,42,43]. $\omega^l_\ell = \omega_p / [l(l+1)]^{1/2}$, and, in particular, to the dipole ($l = 1$) plasmon frequency $\omega^l_1 = \omega_p / \sqrt{3}$, the value known as the giant Mie resonance frequency.

Plasmon oscillations are always damped [see drawings (c) and (d) in Figs. 2 and 3] due to radiation losses and all the relaxation processes included in the relaxation rate $\gamma$. With increasing size, the damping rates $\omega^l_\ell(R)$ initially decrease starting from the values $\omega^l_\ell = \omega^l_\ell(0) = \gamma/2$ in each plasmon mode. If $\gamma$ accounts for electronic relaxation processes leading to dissipation (absorption) of energy in the metal, $\omega^l_\ell$ accounts for the decay of the SP oscillations due to dephasing of the collective electron motion which is often assumed to be statistically “memory destroying”. This quantity has been investigated by various experimental methods [4,39,40]. For particles of radii larger than $R_{\text{min}}$, the size dependence of the total damping rates $\omega^l_\ell(R)$ is due to the size dependence of the radiative damping rates $\gamma^l_\ell(R)$. Whereas radiative damping and energy dissipation are uncorrelated processes, the total damping rate $\omega^l_\ell(R)$ can be written as a sum

$$\omega^l_\ell(R) = \gamma^l_\ell(R) + \gamma^\text{diss}_l, \quad l = 1,2,3,\ldots, \quad (7)$$

where $\gamma^\text{diss}_l = \gamma/2$ for any multipolar mode $l$. Size dependence of the rate $\omega^l_\ell(R)$ results from $\gamma^l_\ell(R)$ size depend-
ence. The initial increase of $\omega_l(R)$ with particle size is followed by the subsequent decrease for sufficiently large spheres as it is illustrated in Figs. 2(c), 2(d), 3(c), and 3(d) for the dipole mode damping $\gamma_l(R)$ for silver and gold nanospheres, respectively.

The optical properties of the surrounding medium can introduce a significant alteration in the multipolar SP resonance frequencies and damping rates for a particle of given radius. In particular, an increase in the optical density of the surroundings introduces an important “red shift” of the plasmon resonance frequencies $\omega_l$, as it is illustrated in Figs. 2, 3(a), and 3(b).

3. Properties of plasmonic nanoantennas in terms of multipolar SP resonance frequencies and plasmon oscillation damping rates

The derived SP size characteristics can be used to tune the absorbing or emitting properties of the plasmonic spherical nanoantennas. Knowing both the multipolar SP resonance frequencies $\omega_l(R)$ and corresponding damping rates $\gamma_l(R)$, one can predict properties of plasmonic particle in response to the external EM field and find the optimal size of a nanosphere for a chosen application. It can be, for example, an effective coherent coupling with some neighbouring particles in a metal nanoparticle array, an enhancement of the EM field near the sphere surface (used, e.g., for SERS spectroscopic technique) or a modification of the far field scattering properties.

Resonant excitation of SP oscillations is possible when the frequency of the incoming light $\omega_{in}$ approaches an eigenfrequency of a plasmonic nanoantenna of the given radius $R$, $\omega_{in} = \omega_l(R)$, $l = 1, 2, 3, \ldots$ [Figs. 2, 3(a), and 3(b)]. If excited, plasmon oscillations are damped at the corresponding rates $\gamma_l(R)$. The sphere acts, then, as a receiving or/and emitting multipolar EM nanoantenna in the mode(s) $l$. The nanoantennas performance can be adjusted by changing the sphere size. Not only the resonance frequencies $\omega_l(R)$ (with $l = 1$ for the dipole antenna mode), but also receiving (absorbing) and scattering (transmitting) abilities of plasmonic nanoantennas are size dependent as follows from the SP damping rates $\gamma_l(R)$ size dependence [Figs. 2, 3(c), and 3(d)]. It is convenient to keep expressions for $\omega_l$, and $\gamma_l$ in (eV) units in the discussion, but for completeness we give the corresponding magnitudes in (1/s) ones on the left vertical axis of Figs. 2, 3(c), and 3(d).

Fig. 2. (a) and (b) multipolar plasmon resonance frequencies $\omega_l(R)$ and (c) and (d) plasmon damping rates $\gamma_l(R)$ ($l = 1, 2, \ldots 7$) for silver nanoparticles in free space ($n_{out} = 1$) and in a suspension ($n_{out} = 1.5$) as a function of sphere radius.

Fig. 3. (a) and (b) multipolar plasmon resonance frequencies $\omega_l(R)$ and (c) and (d) plasmon damping rates $\gamma_l(R)$ ($l = 1, 2, \ldots 7$) for gold nanoparticles in free space ($n_{out} = 1$) and in a suspension ($n_{out} = 1.5$) as a function of sphere radius.
3.1. Receiving and transmitting abilities of plasmonic nanoantenna as an intrinsic property of a plasmonic particle

It is widely known, that optical properties of a perfect-metal spherical particle, which is much smaller than the wavelength of incoming light, are mainly due to the giant absorption at the resonance frequency $\omega_{1}/\sqrt{3}$. That value corresponds to the SP dipole resonance frequency $\omega_{0,l}(R)$ given by Eq. (5) for $l = 1$. However, a plasmonic particle can act as an efficient absorbing antenna not only in the dipole, but also in larger polarity modes for particles of larger sizes. Moreover, in some ranges of the parameters $R$ and $l$, the receiving and radiative abilities of a plasmonic nanoantenna may be comparable. In such a case, the plasmonic fraction in the total extinction spectrum would be manifested by comparable contributions of the absorption and scattering. Scattering and absorbing abilities of spherical nanoantennas can be conveniently described by the relative contribution of the radiative and nonradiative damping rates to the total damping rate $\omega_{n}^{\text{out}}[R]$ [Figs. 2(a)(b) and 3(a)(b)]. Plasmonic nanoparticles can act as the absorbing antennas when the contribution of the nonradiative damping rate in the total one is large. That is possible for particles of some radii $R$, such that $\omega_{n}^{\text{out}}[R] = \omega_{0,l}^{\text{rad}} = \gamma_{\text{diss}}$. In such case, the SP resonance manifestation in the absorption spectrum is expected to be well pronounced, with maxima at the resonance frequencies $\omega_{0,l}(R)$, $l = 1, 2, ...$. It also contributes considerably to the extinction spectrum regardless of how large the particle is. On the other hand, if contribution of the radiative damping $\gamma_{\text{rad}}^{\text{out}}(R)$ to the total SP damping rate $\omega_{n}^{\text{out}}[R]$ is large in comparison with the nonradiative damping $\gamma_{\text{diss}}$, $\omega_{n}^{\text{out}}[R] = \gamma_{\text{rad}}^{\text{out}}(R)$, a particle of the radius $R$ is able to act as an efficient scattering antenna. The range of particle sizes which fulfills these conditions results from the size characteristics of $\omega_{n}^{\text{out}}[R]$ presented in Figs. 2(c)(d) and 3(c)(d) for silver and gold plasmonic nanospheres, respectively. The basic, intrinsic properties of a particle described by the size characteristics of $\omega_{n}^{\text{out}}(R)$ and $\omega_{n}^{\text{out}}[R]$ are reflected in a way the particle responds to the light field.

3.2. SP resonance manifestation in optical signals

Let us discuss some examples of light elastically scattered by gold nanoparticles of chosen sizes in environment with $n_{\text{out}} = 1.5$. According to Mie theory [30–33,44], the optical response to the incoming EM wave can be described in spherical coordinates as square of sum of the partial waves of the TM ("electric") and the TE ("magnetic") EM contributions. Therefore, the fields contributing to the light intensity are inevitably composed of both TM and TE components of different polarities $l$, while the electromagnetic fields of SP dispersion relation [Eq. (1)] are transverse magnetic ones. Constructive and destructive interference of TM and TE components influences the manner of plasmons manifestation in the scattered light intensity (irradiance). For given particle size, the manifestation of the SP resonance depends on the observable quantity, observation angle and polarization geometry. Figure 4 illustrates the spectra of the total scattering $\sigma_{\text{scat}}(\omega)$, extinction $\sigma_{\text{ext}}(\omega)$ and absorption $\sigma_{\text{abs}}(\omega)$ cross-sections for gold spheres with radii 10 nm, 75 nm, and 130 nm [Figs. 4(a), 4(b), and 4(c)], respectively calculated within Mie theory for a suspension ($n_{\text{out}} = 1.5$) of mono-sized gold spheres as an example. For $R = 10$ nm [Fig. 4(a)], the dipole plasmon resonance at $\omega_{1}^{\text{dip}}(R = 10$ nm) becomes apparent mainly in the enhanced absorptive properties of a nanosphere (the maximum in $\sigma_{\text{abs}}(\omega)$ dependence, grey line in Fig. 4(a), due to the large contribution of the nonradiative damping to the total damping rate $\omega_{n}^{\text{out}}[R = 10$ nm] $= \gamma_{2}^{\text{diss}} = 0.036$ eV. The on-resonance enhancement factor is of three orders in magnitude. The peak in the total scattering cross-section $\sigma_{\text{scat}}(\omega)$ is by a factor of ten smaller due to the poor radiation damping $\omega_{\text{rad}}^\text{n}[R = 10$ nm] $\geq \gamma_{\text{rad}}^{\text{out}}(R)$ [Fig. 3(d)]. Therefore, the optical properties (including colours) of such metallic nanoparticles are dominated by the resonant nonradiative plasmon energy dissipation at the dipole SP eigenfrequency $\omega_{1}^{\text{dip}}(R = 10$ nm) [see Fig. 3(b)]. The mentioned above also applies to the absorptive properties of atoms or molecules associated with the resonant (dipole) transition to the short-living excited states of lifetimes of the order of picoseconds. The full
Plasmonic abilities of gold and silver spherical nanoantennas in terms of size dependent multipolar resonance...

width at half maximum \( \Gamma \) (FWHM) of a narrow, Lorentzian-like peak in the total absorption cross-section \( \sigma_{abs}(\omega) \) [Fig. 4(a)] is well described by the plasmon damping rate \( \omega_{rad}^{\prime}(R=10 \text{ nm}) \) see Fig. 3(d)

\[
\Gamma(R=10 \text{ nm}) = 2\omega_{rad}^{\prime}(R=10 \text{ nm}) = \gamma . \tag{8}
\]

The corresponding plasmon damping time \( \tau = 18.3 \text{ p sec} \).

Scattering effects become important for nanoparticles of larger sizes, as known from Mie work [30]. Figure 4(b) illustrates this effect for sphere with the radius \( R = 75 \text{ nm} \). The magnitude of the total scattering cross-section \( \sigma_{scat}(\omega) \) is bigger than that of the total absorption cross-section \( \sigma_{abs}(\omega) \) in the full optical range. The scattering spectrum is broadened and composed of some partially overlapping maxima which are blue shifted in respect to the SP multipolar resonance frequencies \( \omega_{j}(R) \). Using again the SP damping rate characteristics, one can predict the multipolar plasmonic contribution to the scattering.

The collectively oscillating electrons at the resonance frequency \( \omega_{j}(R=75 \text{ nm}) = 1.6 \text{ eV} \) lose their energy mainly due to radiation, the effect accounted in the size augmented radiation rate contribution to the total damping rate: \( \omega_{rad}^{\prime}(R=75 \text{ nm}) = 0.49 \text{ eV} = \gamma_{rad}^{\prime} \) [Fig. 3(d)]. That is why the fraction of the dipole plasmonic absorption is negligibly small. Corresponding dipole damping rate [Fig. 3(d)] is dominated by the radiation damping \( \gamma_{rad}^{\prime}(R=75 \text{ nm}) \geq \gamma_{diss}^{\prime} = 0.036 \text{ eV} \). Therefore, if the plasmon oscillations are continuously excited by light at the resonance frequency of a dipole plasmon \( \omega = \omega_{rad}^{\prime}(R=75 \text{ nm}) \), a sphere is able to efficiently scatter light through the plasmonic mechanism and to work as an excellent emitting antenna at that frequency. Enhancement of the scattered near and far field in the space around the particle is then possible.

The larger is the rate \( \gamma_{rad}^{\prime}(R) \) in comparison with the nonradiative damping rate for successive \( I = 1,2,3, \ldots \), the higher is the peak in the scattering spectrum \( \sigma_{scat}(\omega) \) due to the plasmonic contribution, as demonstrated in Figs. 4(b) and 4(c). But also, the larger is the rate \( \gamma_{rad}^{\prime}(R) \), the larger is the spectral bandwidth of the SP participation in the scattering spectrum around the SP resonance frequency \( \omega_{j}(R) \).

As illustrated in Figs. 4 and 6(b), the larger is the spectral bandwidth of the SP contribution of polarity \( l \) to the spectrum [defined by the rate \( \gamma_{rad}^{\prime}(R) \)], the stronger is the blue shift of the maximum in respect to the SP resonance frequency \( \omega_{j}(R) \). As illustrated in Figs. 4(b) and 4(c), all the large-bandwidth maxima in the scattering spectrum suffer from this effect and are blue-shifted in respect to the SP resonance frequencies \( \omega_{j}(R) \). This effect is due to the important changes with frequency in the imaginary part of the index of refraction \( n_{rad}(\omega) \) [Fig. 6(c)] that affects the frequency dependence of the nonplasmonic contribution (specular reflection) to the scattering. But also, the larger is the spectral bandwidth of the maximum, the more important is the interference effects (constructive and destructive) of the partial TM and TE waves.

Even large plasmonic particles (such as those of the radius \( R = 75 \text{ nm} \) or larger [see Figs. 4(b) and 4(c)]) can act as receiving antennas producing narrow, well pronounced peaks in the absorption (and extinction) spectra, if only the participation of the energy dissipation rate \( \gamma_{diss}^{\prime} \) in the total plasmon damping rates \( \omega_{j}^{\prime}(R) \) in the mode \( I \) is comparable with the radiative rate \( \gamma_{rad}^{\prime}(R) \) [see Eq.(7)]. The well pronounced maxima in the total absorption cross-sections \( \sigma_{abs}(\omega) \) (black lines in Fig. 4) near the plasmon frequencies \( \omega_{j=2,3}(R=75 \text{ nm}) \) [Fig. 4(b)] and \( \omega_{j=4}(R=130 \text{ nm}) \) [Fig. 4(c)] are some examples. The well pronounced absorption peaks due to SPs are not affected by shifting and smearing out effects suffered by maxima in the scattered spectra, while the SP absorption is due to the plasmonic dependent energy dissipation only. Therefore, interference of EM waves does not affect the position and width of maxima in \( \sigma_{abs}(\omega) \) (black lines in Fig. 4). The contribution of the dipole SP resonance to the maximum in the scattering spectrum of a sphere with \( R = 130 \text{ nm} \) is smaller than for a sphere with \( R = 75 \text{ nm} \), as one can expect knowing that \( \omega_{j=4}(R=130 \text{ nm}) < \omega_{j=1}(R=75 \text{ nm}) \) [see Fig. 3(d)]. Such relation results from the smaller contribution of the radiative damping in the total SP damping rate \( \gamma_{rad}^{\prime}(R=130 \text{ nm}) < \gamma_{rad}^{\prime}(R=75 \text{ nm}) \).

As it was just mentioned, the partial smearing out and blue shift of the maxima in the scattering (extinction) spectra is due to the interference of EM fields scattered by a sphere. Such fields are inevitably composed of both the TM and TE components of the different polarity \( l \), while the resonant SP’s contributions are due to the normal to the sphere surface component of the electric field \( E_{\perp} \), that is present in the TM polarization mode only. The TE fraction of eddy currents is also size dependent and increases monotonically with \( R \), contributing also at some amount to the blue shift of the maxima in the total scattering cross-section [29].

One can conclude, that if the SP damping rates \( \omega_{rad}^{\prime}(R) \) for \( I = 1,2, \ldots \) of a plasmonic active sphere of the radius \( R \) are mainly due to the nonradiative damping \( \omega_{rad}^{\prime}(R) = \gamma_{diss}^{\prime} \) [Fig. 3(d)], such sphere is able to efficiently absorb light through the plasmonic mechanism near the corresponding SP resonance frequency \( \omega_{j}^{\prime}(R) \) in the spectrally narrow bandwidth \( \gamma_{diss}^{\prime} \) defined by the relaxation rate \( \gamma \). However, such receiving antennas are rather unable to couple electromagnetically with another particle by the plasmonic mechanism.

If the SP damping rate \( \omega_{rad}^{\prime}(R) \) is much larger than \( \gamma_{diss}^{\prime} \), a plasmonic particle of the size \( R \) is a good radiating antenna at the resonance frequency \( \omega_{j}(R) \) of the given polarity \( l \). Radiative damping, which is inherently coupled to plasmon oscillations, affects not only the spectral width of the plasmon related maxima in the scattered light intensity, but also enhances the plasmon related contributions to the scattered spectrum. For the large radiation damping rates \( \omega_{rad}^{\prime}(R) \), the energy of the incident EM wave can be effectively redistributed into the scattered field energy around the sphere due to the plasmonic mechanism (a sphere acts as a radiating antenna).
3.3. Plasmonic scattering in orthogonal polarization geometries

A distinct dipole and quadrupole scattering abilities of the plasmonic nanoantenna can be conveniently demonstrated by linearly polarized light illuminating the plasmonic sphere at the right angle to the direction of the incoming light beam (Fig. 5).

Such “clinical” orthogonal scattering geometry enables to expose a contribution of both: a single dipole ($l = 1$) and a single quadrupole ($l = 2$) SP to the scattering signals and to separate these contributions spatially by observing $I_\perp(\omega, R)$ and $I_\parallel(\omega, R)$ intensities (Fig. 4) [29,45]. Whereas $I_\perp$ and $I_\parallel$ are the intensities of the purely scattered light, while there is no influence of the interference effect with the EM field of the incoming light wave from the perpendicular direction. Therefore, such observation enables to study pure scattering abilities of a plasmonic sphere. Using such an experimental geometry and observing the changes in $I_\perp(R)/2\pi R^2$ and $I_\parallel(R)/2\pi R^2$ with size [46] we have studied the size dependence of the dipole and quadrupole plasmon resonances in our experiment on spontaneously growing sodium droplets (up to the droplet radius $R = 150$ nm) induced by laser light [28,29]. Studying the intensity scattered by the particle unit area allowed us to diminish a contribution of the background due to eddy currents and to emphasize SP manifestation.

The changes in the spectra of $I_\perp(\omega)$ and $I_\parallel(\omega)$ with particle radius, discussed in terms of the intrinsic SP multipolar size characteristics (Sect. 2), can directly illustrate the role of the SP radiative damping rates $\gamma_\perp(R)$ in formation of the radiative properties of the plasmonic nanoantennas.

The FWHM $\Gamma_\text{scat}$ of the $I_\perp(\omega)$ spectrum [Figs. 6(a) and 6(b), solid line], and $\Gamma_\text{scat}$ of the $I_\parallel(\omega)$ spectrum [Figs. 6(a) and 6(b), dashed line] can be described by the Lorentzian function of FWHM corresponding values of the FWHM

$$\Gamma_\text{scat}^\parallel(R) = 2[a_\parallel(R)].$$

The peaks in the spectra for spheres of larger sizes become blue shifted in respect to the intrinsic value $\omega^{\text{scat}}_{l=2}(R)$ of the SP dipole and quadrupole SP resonances, as illustrated in Figs. 6(a) and 6(c). The larger the spectral width $\Gamma_\text{scat}$, the stronger the blue shift of the maximum position $\omega^{\text{scat}}_l(R)$ in respect to the SP plasmon resonance position $\omega_l(R)$, as illustrated in Fig. 6(b). The maxima due to SP resonances in the scattering spectra are governed by the radiative damping rates $\gamma^{\text{rad}}_l = |\omega_l(R)| - \gamma^{\text{diss}}_l$ for dipole ($l = 1$) and quadrupole ($l = 2$) plasmon mode, correspondingly.

One can conclude that changing the polarization geometry it is possible to separate the quadrupole plasmon radiation from the dipole and from the larger multipolarity plasmon contribution to the scattered light. Knowing the manner of dipole and quadrupole plasmon manifestation in $I_\perp(\omega)$ and $I_\parallel(\omega)$ spectra correspondingly, and $\omega^{\text{scat}}_{l=2}(R)$ and $\omega^{\text{scat}}_{l=2}(R)$ size dependencies, it is possible to switch between the receiving (enhanced absorption) and emitting (enhanced scattering) abilities of a plasmonic nanoantenna by changing the direction of observation (Fig. 5) or direction of polarization in respect to the observation plane. It is also possible to control a bandwidth of particle extinction or scattering spectra changing it from spectrally broad to spectrally narrow and vice versa, by changing the wavelength [see Fig. 4(b) or Fig. 6(b) as an example] of the illuminating light, if a particle is sufficiently large.

4. Conclusions

Receiving or/and emitting properties of the spherical plasmonic nanoantenna are governed by the inherent parameters, the size dependent plasmon resonance frequencies $\omega_l(R)$ and
damping rates $\kappa_\gamma(R) = \gamma_1^l (R) + \gamma_{\text{diss}}^l$, which provide a complete description of particle plasmonic properties. Knowledge of the size dependence of both, the multipolar SP resonance frequencies and corresponding damping rates makes possible to effectively describe particle plasmonic properties in response to the EM field. Taking advantage of the particle intrinsic size characteristics it is possible to predict not only the SP resonance frequencies $\omega_\gamma(R)$ but also the strength of SP resonances in given polarity order $l = 1, 2, 3,...$ after proper adjustment of the relative magnitude of the radiative $\gamma_1^l (R)$ and energy dissipative $\gamma_{\text{diss}}^l$ damping rates.

If the SP multipolar damping rate $\kappa_{\omega_\gamma}^l(R)$ of a plasmon active sphere of the radius $R$ is mainly due to the nonradiative rate $\kappa_{\omega_\gamma}^l(R) = \gamma_{\text{diss}}^l = \gamma / 2$ [Fig. 3(d)], such sphere is able to efficiently absorb light through the plasmonic mechanism near the corresponding SP resonance frequency $\omega_\gamma(R)$ in the spectrally narrow bandwidth $\gamma_{\text{diss}}^l$, even for larger sizes (and larger $l$). However, such receiving antenna is unable to couple electromagnetically with another particle by the plasmonic mechanism. If the SP damping rate $\kappa_{\omega_\gamma}^l(R)$ is much larger than $\gamma_{\text{diss}}^l$, a plasmonic particle of the corresponding size is a good radiating antenna at the resonance frequency $\omega_\gamma(R)$ and is able to enhance the EM field in far and near field region.

Using intrinsic plasmon size characteristics $\omega_\gamma(R)$ and $\omega_{\omega_\gamma}^l(R)$ it is possible to control an operation resonance frequency of plasmonic nanoantenna and change the operation range from spectrally broad to spectrally narrow and vice versa [large or small $\omega_{\omega_\gamma}^l(R)$]. It is also possible to switch between the particle receiving (enhanced absorption) and emitting (enhanced scattering) abilities in the qualitatively controlled manner. Changing the polarization geometry (Sect. 3.2) it is possible to effectively separate the dipole or quadrupole plasmon radiation from all the non-plasmonic contributions to the scattered light.

Acknowledgements

We would like to acknowledge support of this work by the Polish Ministry of Science and Higher Education under Grant No N N202 126837.

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