What we expect from weakly dissipating materials at the range of plasmon resonance frequencies

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***Abstract* - Development of modern materials, including nanoclusters, cluster assembled materials and metamaterials is among the actual challenges for the development of future nanotechnologies. Here we discuss the peculiarities of far-field and near-field light scattering by plasmonic nanoparticles, and possible applications of weakly dissipating materials. Over the last few years many peculiarities of light scattering have been found for nanoparticles in the regime of plasmon resonances. Optical excitation of localized plasmons is accompanied by inverse process - transformation of localized resonant plasmons into scattered light. When radiative damping prevails over the dissipative damping, the effects of anomalous light scattering result in sharp giant optical resonances and complicated near-field structure of the Poynting vector field, see e.g. [1-4]. Here we present peculiarities of far-field and near-field light scattering by plasmonic nanoparticles with weak dissipation and anisotropy.**

Lord Rayleigh [5] performed the first analysis of light scattering by small scatters and found formulas for a spherical particle and thin cylinder (Rayleigh scattering). Later it was shown that these formulas are certain limits of the exact solution of Maxwell equations, obtained by Mie [6] for spherical particles and of a similar solution for a thin wire [7]. These formulas however contain resonance denominators. For weakly dissipating materials it results in unreasonably large scattering cross-sections at the plasmon resonance frequencies, which is explained by violation of the applicability conditions for the Rayleigh approximation [1-4,8]. The thorough analysis of the exact Mie solution in this case reveals a number of unexpected features of the light scattering, such as giant optical resonances with an inverted hierarchy (the quadrupole resonance occurs more intensively than the dipole, etc.), a complicated near-field structure with vortices, and unusual frequency and size dependencies of the scattered light [1-4,8]. Probably, the most intriguing feature is related to the great sensitivity of the Poynting vector field to small variations in light frequency, see e.g. Fig. 1, where transforms of the field in the vicinity of dipole resonance are shown. With sufficient deviation from the dipole plasmon resonance frequency (or with sufficiently high dissipation) one returns back to the field distribution typical of the Rayleigh scattering [9]. This provides a surprisingly powerful tool for manipulating with energy flows at nanoscales with help of the weakly dissipating materials.

An even more fascinating effect was found in the vicinity of quadrupole resonance, where extra high sensitivity of the angular distribution of scattering light can be seen [10, 11]. A very small variation in the incident light frequency changes the scattering diagram from forward scattering to backward scattering, as shown in Fig. 2. Recently, it has been revealed [12] that this effect occurs identical to the well-known Fano resonances in quantum physics [13]. In this case the localized plasmons (polaritons), exited by the incident light in the scattering particle, are equivalent to the quasi-discrete levels in the Fano approach, while the radiative decay of these excitations plays exactly the same role as tunneling from the quasi-discrete levels in the quantum problem. As a result the resonance may have a typical N-shaped line with a local maximum, corresponding to the constructive interference of different eigenmodes and a local minimum, corresponding to the destructive one. In particular, the destructive interference may result in considerable, or even complete suppression of the scattering along any given direction. In the vicinity of the dipole resonance we always get a Lorenzian scattering contour [1], while in the vicinity of the quadrupole resonance an asymmetric Fano resonance profile [12] may be observed. Thus, the famous Fano resonance was, in fact, have been hidden in the exact Mie solution [14].

We also discuss light scattering by a spherical particle with radial anisotropy by extending the Mie theory to the diffraction by an anisotropic sphere, including both electric and magnetic anisotropy ratio [16, 17]. It is shown that radial anisotropy may lead to great modifications in scattering efficiencies and field enhancement, elucidating the importance of anisotropies in control of scattering. Even a small variation in the anisotropy plays an important role in enhancing or suppression of scattering efficiencies, see in Fig. 3 a, b.

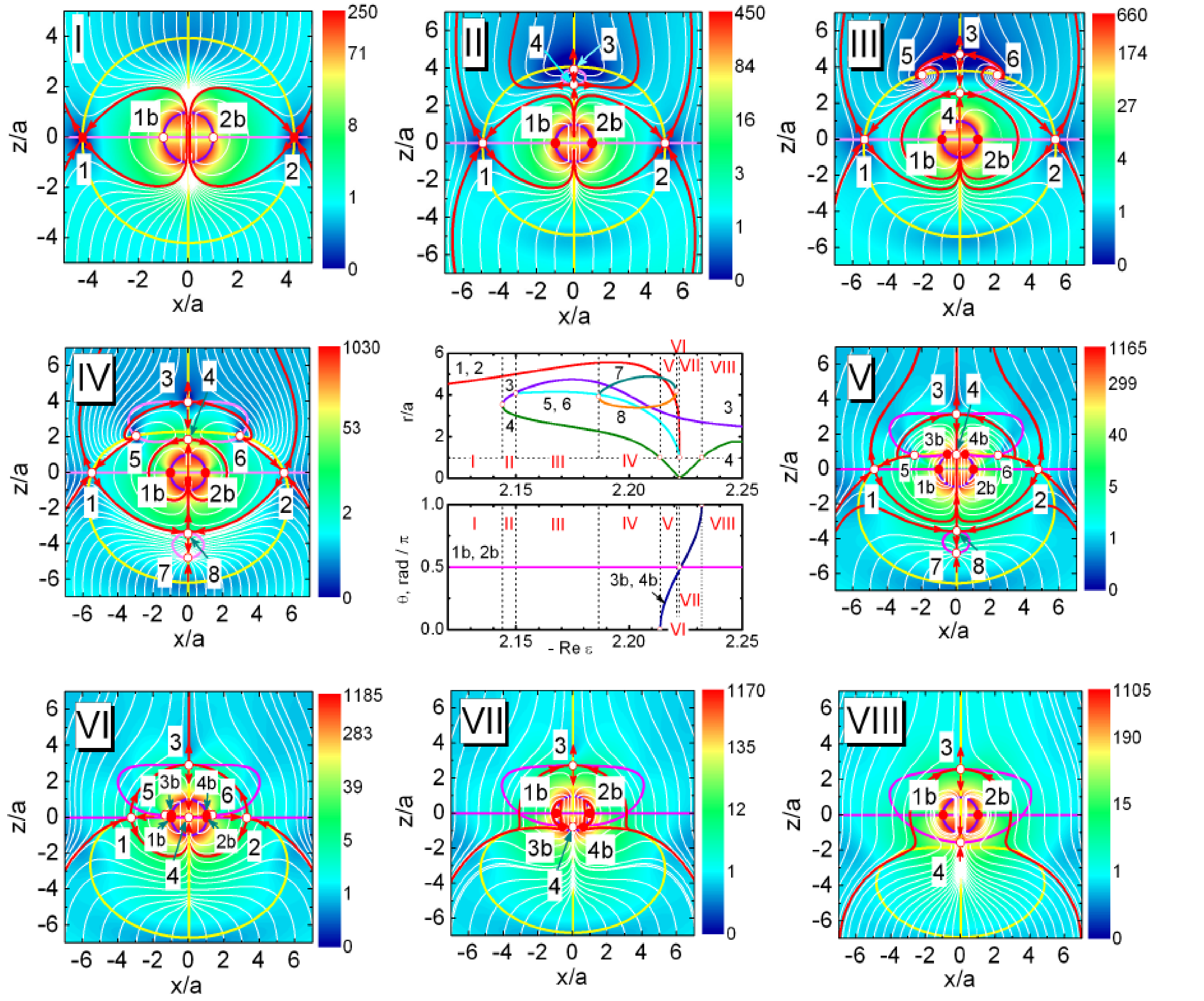

Fig. 1. The central panel presents the distances from the particle center to various singular points (marked with italic Arabic numerals) of the Poynting vector field as a function of dielectric function ε in the vicinity of the dipole resonance at size parameter $q = 2\pi a/\lambda = 0.3$, and the corresponding field distributions at the characteristic values of ε. Roman numerals designate regions with fixed topological structures of the field. Color density plots show values of the modulus of the Poynting vector $\mathbf{S}$ (log scale) normalized over its value for the incident light. Field lines are shown in white, separatrices in red, null-isoclines $S_\theta(r,\theta)=0$ in yellow, $S_r(r,\theta)=0$ in pink, the particle surface in violet, and singular points as open red circles. Note two optical vortices in panel III (Arabic numerals 5 and 6). Three of these panels (I, III and VIII) were presented previously in [1].

Similar phenomena may be observed in the field distribution. However, it should be emphasized that these effects are sharply suppressed with an increase in the dissipation rates. The Fano resonance may also be enhanced by the anisotropy, see in Fig. 3 c.

We also discussed the acoustic phenomena initiated by excitation of plasmon resonances in coupled plasmonic structures and the effects of Surface Enhanced Raman Scattering [18, 19]. We investigated optoacoustic effects in the system of coupled plasmonic nanospheres illuminated simultaneously by a laser and acoustic waves [20]. Calculations show that within this system one can control the acoustical band gap at low frequency as well as enhance greatly the acoustic signal at resonant frequencies, as shown in Fig. 4.

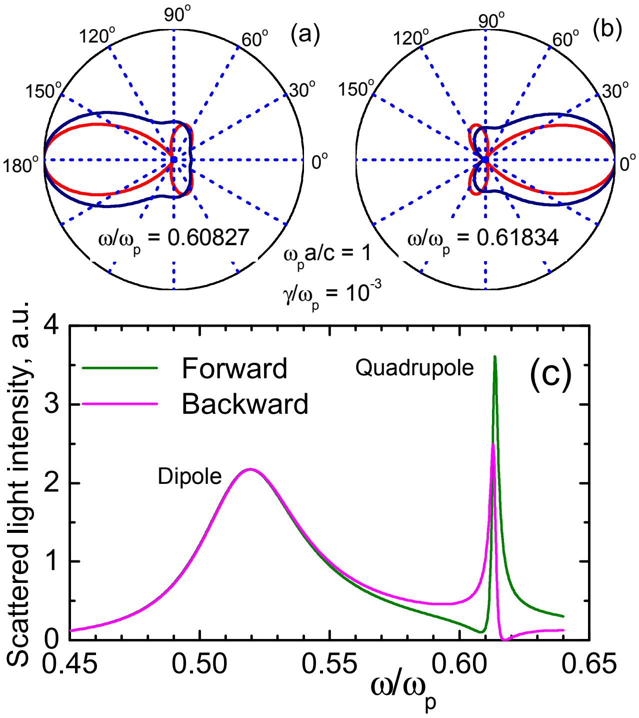

Fig. 2. The angular dependence of light scattering that follows from the exact Mie solution. The dielectric function was described by the Drude formula with weak dissipation $\gamma/\omega_p = 10^{-3}$. Radius of the particle, a, at the dipole resonance is significantly smaller then the corresponding wavelength λ, $a/\lambda \approx 0.083$. Scattering diagrams in (a) and (b) are defined by the standard manner [15] for linearly polarized (red lines) and nonpolarized (blue lines) radiation. Note the transformation of the diagram from forward to backward scattering coursed by a fine detuning of the frequency in the vicinity of the quadrupole resonance. One can also see the asymmetric forward (olive) and backward (magenta) scattering profiles associated with the Fano resonance (c).

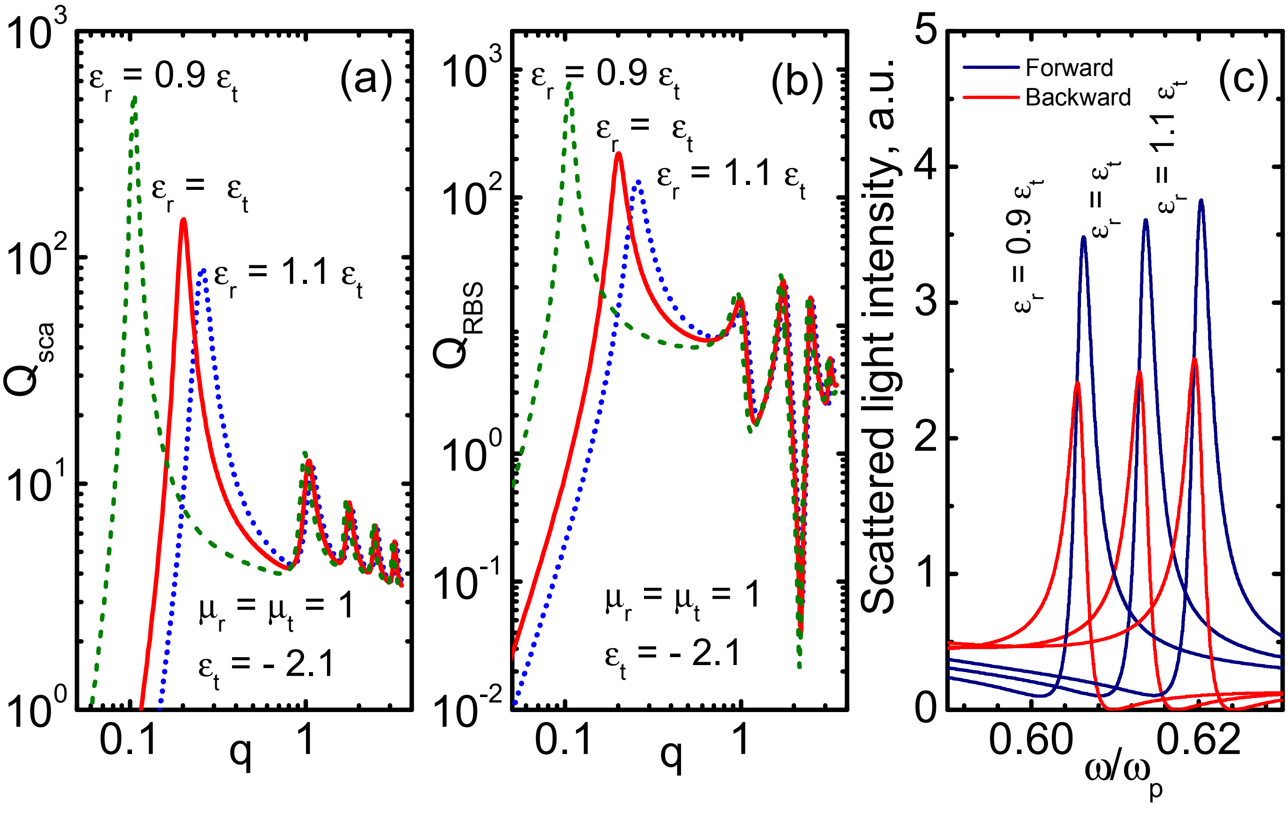

Fig. 3. (a) Scattering efficiencies associated with the dipole surface plasmon resonances versus size parameter $q = 2\pi a/\lambda$ for nondissipating anisotropic materials with different radial ε_r and transversal ε_t components of permittivity. (b) Radar backscattering signal variation for an anisotropic material. (c) Variation of the Fano resonances within an anisotropic material with weak dissipation rate $\gamma/\omega_p = 10^{-3}$.

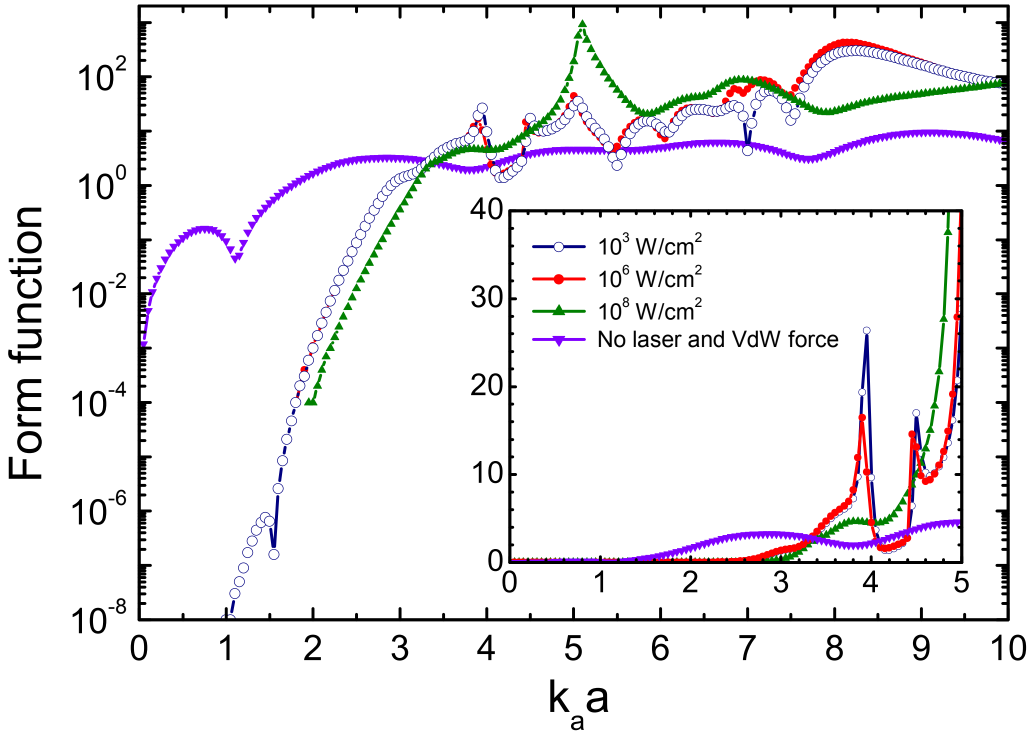

Fig. 4. Computed acoustic form faction [20] of two elastic a = 60 nm - sized Ag nanospheres at separation distance 1 nm, illuminated by a 514 nm laser with different intensities, versus acoustic size parameter $k_a a$ (k_a is acoustic wave number). In addition to the elastic forces, the optical force induced by light pressure and Van der Waals force are taken into account.

This study shows that weakly dissipating materials may have many attractive applications in plasmonics and optoacoustics. They provide new opportunities for a giant, controlled, highly frequency-sensitive enhancement and variation of electromagnetic field at nanoscales.

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