Light scattering at nanoparticles close to plasmon resonance frequencies

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(Submitted December 12, 2005)
Opticheskiĭ Zhurnal **73**, 7–14 (June 2006)

This paper discusses light scattering by small particles in the neighborhoods of plasmon resonances in the small-dissipation limit. The concept that the scattering has a dipole character (Rayleigh scattering) cannot be used in this limit, and the corresponding formulas must be replaced by others, in which the main role is played by effects associated with radiation losses caused by the transformation of localized plasmons into scattered radiation. *© 2006 Optical Society of America*.

INTRODUCTION

In speaking of the scientific school of Alekseĭ Mikhaĭlovich Bonch-Bruevich, you involuntarily think that this school long ago reached the formal limits of his direct students. *Cui bono*—for whose benefit? Most of those who were fortunate enough to participate in the all-Union conferences on nonresonance interaction of optical radiation with matter organized by Alekseĭ Mikhaĭlovich have the right to consider themselves at least partly representatives of this school. The history of science establishes in retrospect the quality of a scientist's intuition, his capacity of "being ahead of his time." In publishing this article in A. M. Bonch-Bruevich's birthday collection, we recall with gratitude our numerous discussions, and we are struck by the fact that many questions discussed decades ago at those conferences still remain crucial.

Among the problems in the range of scientific interests of Alekseĭ Mikhaĭlovich and his students is that of the excitation of plasmons in metals under laser stimulation and its various applications (see, for example, Refs. 1–8). This problem has acquired special interest in recent years in connection with the development of nanotechnologies.[9,10] Even though light scattering by a small particle is among the fundamental problems of electrodynamics,[11,12] the physical understanding of the process is still based on the concepts developed by Lord Rayleigh in 1871.[13] Namely, it is assumed that a small particle behaves like an electric dipole—for example, a point dipole in the case of a small spherical particle or a linear dipole in the case of a thin wire.

It is actually assumed in classical electrodynamics that light is scattered as a result of the polarization of the scattering particle by the incident light wave. Because the particle is small, it can be assumed that a light field of such a size is homogeneous. Since a spatially homogeneous field causes only dipolar polarization, a small particle scatters light as a vibrating dipole. The higher orders of scattering—quadrupole, octupole, etc.—are substantially suppressed in this case. For a spherical particle in vacuum, these arguments lead to the well-known formulas for the Rayleigh scattering cross sections:[11,14,15]

$$Q_{\rm sca} = \frac{8}{3}\left|\frac{n^2-1}{n^2+2}\right|^2 q^4 \text{ for a spherical particle} \qquad (1)$$

and

$$Q_{\rm sca} = \frac{\pi^2}{4}\left|\frac{n^2-1}{n^2+1}\right|^2 q^3 \text{ for a thin cylinder.} \qquad (2)$$

The formulas for the cross sections are written in normalized units. To obtain the dimensional scattering cross sections $\sigma_{\rm sca}$, the $Q_{\rm sca}$ values should be multiplied by the geometrical cross section $\sigma_{\rm geom}$. For a sphere, this cross section is $\sigma_{\rm geom}=\pi a^2$, where a is the radius of the particle. In the case of a thin cylinder, $\sigma_{\rm geom}=2aL$, where a is the radius of the cylinder, and $L \gg a$ is the length of the cylinder (the case is considered in which the wave vector of the incident wave is perpendicular to the axis of the cylinder, while vector $\mathbf{H}$ is parallel to it). The quantity $q=2\pi a n_m/\lambda \ll 1$ in Eqs. (1) and (2) is the so-called size parameter, λ is the wavelength of the incident radiation in vacuum, and n_m is the refractive index of the medium in which the particle is submerged. The complex refractive index of the particle itself is determined by the quantity n_p, and the ratio $n=n_p/n_m$ gives the relative change of the refractive index. The relative variation of the permittivity is accordingly determined by the formula $\varepsilon=\varepsilon_p/\varepsilon_m \equiv n_p^2/n_m^2$.

It follows from Eq. (1) that the scattering is proportional to the fourth power of the frequency; as is well known, this explains why the sky is blue. At the same time, Eqs. (1) and (2) contain resonance denominators, which lead to divergences when $n^2=-2$ (for a sphere) and $n^2=-1$ for a thin cylinder). The physical cause of these divergences is also well known. The bounded particle is a resonator that has its

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own modes (localized plasmons). When the frequency of the light coincides with the frequency of the corresponding dipole mode, we have a trivial resonance. When there is no dissipation in the particle, the amplitude of the resonance mode diverges, as in the excitation of vibrations by an external periodic force in an oscillator without friction. Therefore, the ordinary way to avoid divergence in Eqs. (1) and (2) is to take into account dissipative losses, $\mathrm{Im}\,\varepsilon \neq 0$, analogous to what occurs when friction is turned on in the problem of the vibrations of a harmonic oscillator. Since any classical physical system must have some dissipative mechanism, $\mathrm{Im}\,\varepsilon \neq 0$, and Eqs. (1) and (2) give finite values of the scattering cross sections. For all that, a certain discomfort remains for systems with small dissipation. Rayleigh scattering formally allows the possibility that a nanoparticle with small dissipation can have a scattering cross section, for example, of one square kilometer.[1)]

A solution of the paradox of light scattering by a particle with low dissipation was found in Ref. 17. As shown in that paper, along with ordinary dissipation, for which light energy is converted into Joule heat, there is a radiation mechanism of dissipation, associated with losses of plasmon energy, caused by the inverse transformation of a localized plasmon into propagating electromagnetic radiation. The most astonishing circumstance in this case is that the given mechanism is contained in the exact formulas of Mie theory and in principle could have been detected a hundred years ago. The Mie theory in the corresponding limit gives formulas that differ qualitatively from Eqs. (1) and (2). In what follows, we shall call the corresponding effects *anomalous light scattering* close to the plasmon resonance frequencies.

Anomalous light scattering has a number of surprising properties. Some of them, such as the "inverse hierarchy of optical resonances," were detected more than twenty years ago.[17] However, others, associated with the formation of a complex structure of the energy flux in the near-field region (in particular, the formation of optical vortices) were found only recently.[20,21]

In this paper, we discuss a number of features of anomalous light scattering close to the plasmon resonance frequencies.

A LOCALIZED PLASMON AND ENERGY DISSIPATION

We first of all recall how the Rayleigh formulas follow from exact solutions of Maxwell's equations. In the case of a spherical particle, the corresponding solutions are given by Mie theory, which gives the following (exact) expressions for the extinction, scattering, and absorption cross sections:[11]

$$Q_{\mathrm{ext}} = \frac{2}{q^2}\sum_{l=1}^{\infty}(2l+1)\mathrm{Re}(a_l+b_l),$$

$$Q_{\mathrm{sca}} = \frac{2}{q^2}\sum_{l=1}^{\infty}(2l+1)\{|a_l|^2+|b_l|^2\},$$

$$Q_{\mathrm{abs}} = Q_{\mathrm{ext}} - Q_{\mathrm{sca}}. \tag{3}$$

The scattering amplitudes are given by

$$a_l = \frac{n\Psi_l'(q)\Psi_l(nq) - \Psi_l(q)\Psi_l'(nq)}{n\varsigma_l'(q)\Psi_l(nq) - \Psi_l'(nq)\varsigma_l(q)},$$

$$b_l = \frac{n\Psi_l'(nq)\Psi_l(q) - \Psi_l(nq)\Psi_l'(q)}{n\Psi_l'(nq)\varsigma_l(q) - \Psi_l(nq)\varsigma_l'(q)}. \tag{4}$$

Here we use the same notation as in Eq. (1). Functions $\psi_l(z)=\sqrt{\pi z/2}J_{l+1/2}(z)$ and $\varsigma_l(z)=\sqrt{\pi z/2}H_{l+1/2}^{(1)}(z)$ represent the spherical Bessel and Ricatti-Bessel functions. Here $J_v(z)$ is the Bessel function, $H_v^{(1)}(z)=J_v(z)+iN_v(z)$ is the Hankel function (a Bessel function of the third kind), where $N_v(z)$ is the Neumann function (a Bessel function of the second kind, sometimes denoted by $Y_v(z)$]. The primes in Eqs. (4) denote derivatives with respect to the argument; i.e., $\psi_l'(z) \equiv d\psi_l(z)/dz$, etc.

Using the expansion of the functions in small q, it can be shown that, for nonmagnetic media, $a_l \gg b_l$, and therefore the terms containing b_l in Eqs. (3) can be omitted. The amplitude of a_l can be conveniently represented in the form

$$a_l = \frac{\Re_l}{\Re_l + i\Im_l}, \tag{5}$$

where functions $\mathcal{R}_l$ and $\mathcal{J}_l$ are determined by

$$\Re_l(q) = n\Psi_l'(q)\Psi_l(nq) - \Psi_l(q)\Psi_l'(nq),$$

$$\Im_l(q) = n\chi_l'(q)\Psi_l(nq) - \Psi_l'(nq)\chi_l(q), \tag{6}$$

where $\chi_l(z)=\sqrt{\pi z/2}N_{l+1/2}(z)$. Using the well-known expansions for the Bessel functions for small values of the argument,[20] it is easy to obtain that, for $q \ll 1$,

$$\Re_l(q) \approx q^{2l+1}\frac{l+1}{[(2l+1)!!]^2}n^l(n^2-1). \tag{7}$$

At the same time, the expansion of $\mathcal{J}_l(q)$ for small q begins with a constant term,

$$\Im_l(q) = n^l\frac{l}{2l+1}\left[n^2+\frac{l+1}{l}-\frac{q^2}{2}(n^2-1)\left(\frac{n^2}{2l+3}+\frac{l+1}{l(2l-1)}\right)+\dots\right]. \tag{8}$$

Since, for small q, there is the relationship $\mathcal{R}_l = O(q_m^{2l+1}) \ll \mathcal{J}_l = O(1)$, the ordinary approximation for amplitudes a_l consists of neglecting the value of $\mathcal{R}_l$ in the denominators of Eqs. (5). The dipole scattering amplitude in this case is given by[14,15]

$$a_1 \approx -\frac{2i}{3}\frac{n^2-1}{n^2+2}q^3. \tag{9}$$

Substituting Eq. (9) into Eq. (3) for Q_{sca}, we arrive at Rayleigh's formula, Eq. (1), for dipole scattering[2)]

$$Q_{\mathrm{sca}} \approx \frac{6}{q^2}|a_1|^2 = \frac{8}{3}\left|\frac{n^2-1}{n^2+2}\right|^2 q^4. \tag{10}$$

Let us consider the case in which the permittivity of a metal is described by the Drude formula,

$$\varepsilon = n^2 = 1 - \frac{\omega_p^2}{\omega^2 + \gamma^2} + i\frac{\gamma}{\omega}\frac{\omega_p^2}{\omega^2 + \gamma^2}. \qquad (11)$$

Here, as usual, ω_p denotes the plasma frequency, while γ is the frequency of electron collisions. Substituting Eq. (11) into Eq. (10), we get a Lorentz scattering contour,

$$Q_{\text{sca}}^{(\text{Ra})} = \frac{8}{3}\frac{\omega_{\text{sp}}^4}{(\omega^2 - \omega_{\text{sp}}^2)^2 + \omega^2\gamma^2}q^4, \qquad (12)$$

where $\omega_{\text{sp}} = \omega_p/\sqrt{3}$ is the resonance frequency of the surface plasmon in the case of dipole resonance. As can be seen from Eq. (12), the resonance width is directly connected with the parameter γ responsible for dissipation. The Drude formula can be written in a similar way in the absence of dissipation ($\gamma=0$) in the expression for the exact dipole scattering amplitude, $Q_{\text{sca}} \approx 6|a_1|^2/q^2$. We shall use $a_1 = \mathcal{R}_1/(\mathcal{R}_1 + i\mathcal{J}_1)$, where $\mathcal{R}_1$ and $\mathcal{J}_1$ are determined from Eqs. (7) and (8). We should point out that the plasmon resonance frequencies are determined from the condition $\mathcal{J}_l(q)=0$, and therefore, close to the resonance frequency, when $q \ll 1$, we have $\mathcal{J}_1 \approx i\sqrt{2}(\omega^2 - \omega_{\text{sp}}^2)/\omega_{\text{sp}}^2$. In this case, $\mathcal{R}_1 \approx -2i\sqrt{2}q^3/3$. As a result, we again arrive at a Lorentz contour of the scattering close to resonance:

$$Q_{\text{sca}} = \frac{8}{3}\frac{\omega_{\text{sp}}^4}{(\omega^2 - \omega_{\text{sp}}^2)^2 + \frac{4}{9}q^6\omega_{\text{sp}}^4}q^4. \qquad (13)$$

Comparing Eqs. (12) and (13), it is easy to see that the role of the dissipation parameter in Eq. (13) is played by the quantity

$$\gamma_{\text{eff}} = \frac{2}{3}\omega_{\text{sp}}q^3 = \frac{2}{3}\frac{\omega_{\text{sp}}^4 a^3}{c^3}. \qquad (14)$$

Here c is the velocity of light, and $q = \omega a/c$. This dissipation is caused by the finite plasmon lifetime $\tau_p = \gamma_{\text{eff}}^{-1}$ and the corresponding radiation losses. The effects of a finite plasmon lifetime have already been discussed in the literature. However, they were estimated by another value: $1/\tau_p \approx v_F/a$,[21] where v_F is the Fermi velocity of the electrons. Such a mechanism is caused by the collisions of the electrons with the surface of the particle, as a result of which the plasmon lifetime decreases with decreasing particle size; i.e., $\tau_p \propto a$. In our case, the corresponding time sharply increases as the particle decreases, $\tau_p \propto a^{-3}$. This suggests that the plasmon-radiation mechanism in Mie theory is not at all associated with collisions of the electrons with the surface of the particle.

In the case of exact plasma resonance, when $\omega = \omega_{\text{sp}}$, the Rayleigh formula gives

$$Q_{\text{sca res}}^{(\text{Ra})} = \frac{8}{3}\frac{\omega_{\text{sp}}^2}{\gamma^2}q^4 = \frac{8}{3}\frac{\omega_{\text{sp}}^6 a^4}{\gamma^2 c^2}, \qquad (15)$$

i.e., the scattering efficiency sharply declines with decreasing particle size. Equation (13), however, demonstrates the opposite property,

$$Q_{\text{sca res}}^{(\text{Mie})} = \frac{6}{q^2} = 6\frac{c^2}{\omega_{\text{sp}}^2 a^2}, \qquad (16)$$

i.e., the scattering efficiency increases with decreasing particle size. It can already be seen from this that the anomalous scattering of light has little in common with ordinary Rayleigh scattering. Since all the amplitudes a_l at frequencies of the corresponding plasmon resonances $\omega_l^2 = \omega_p^2 l/(2l+1)$ go to unity, the theory of anomalous scattering of light predicts an inverse hierarchy of the resonances and, in general,[17]

$$Q_{\text{ext}} = Q_{\text{sca}} \approx Q^{(l)} = \frac{2(2l+1)}{q_l^2} = \frac{2}{l}(2l+1)^2\frac{c^2}{\omega_p^2 a^2}. \qquad (17)$$

This means that the quadrupole resonance, $l=2$, at the corresponding frequency has a 40% greater cross section than the dipole resonance, with $l=1$. However, the amplitude of the octupole resonance, $l=3$, exceeds the amplitude of the dipole resonance by 80%. Such an effect exists for nonabsorbing materials, where $\varepsilon'' = \text{Im}\,\varepsilon = 0$. In general, when small energy dissipation in a particle exists and $\varepsilon'' \neq 0$, the Mie theory results in the inequality[17]

$$\varepsilon''(\omega_l) \ll \frac{q_m^{2l+1}}{l[(2l-1)!!]^2}. \qquad (18)$$

Anomalous scattering dominates when this inequality is satisfied, and ordinary Rayleigh scattering is restored when it breaks down, with all the resonances other than dipole strongly suppressed. If $q \to 0$, inequality (18) breaks down for any finite value of ε''. This means that anomalous scattering can be observed, but only for particles that are not too small. An increase of l also causes inequality (18) to break down. This means that the normal Rayleigh hierarchy of resonances is restored beginning with a certain value of l_{max}. The limitation of inequality (18) explains why, despite many years of study of light scattering at nanoparticles (see, for example, Ref. 21), anomalous scattering has not yet been detected experimentally. First, the necessary condition $\varepsilon''(\omega_l) \ll 1$ is not satisfied for most metals that have been studied (gold, silver, and mercury). Second, the inverse hierarchy can be observed only for several of the first resonances $l < l_{\text{max}}$. At the same time, anomalous scattering of light is not a purely theoretical effect, and two suitable candidates for experimental observation were proposed in our earlier papers.[17,21] The first example, indicated in Ref. 17, relates to potassium nanoclusters localized in a matrix of the additively colored alkali halide crystal KCl. The second example[22] relates to aluminum nanoclusters, which also possess a small value of Im ε at the frequency of the first plasmon resonance (at 140 nm). Calculations show that it is possible to observe anomalous scattering for quadrupole resonance in the case of a KCl crystal, and it is even possible to see the first four resonances caused by anomalous scattering on aluminum nanoclusters.

ENERGY FLUX DISTRIBUTION

In order to understand the mechanism of plasmon radiation, one should turn to an investigation of the energy flux

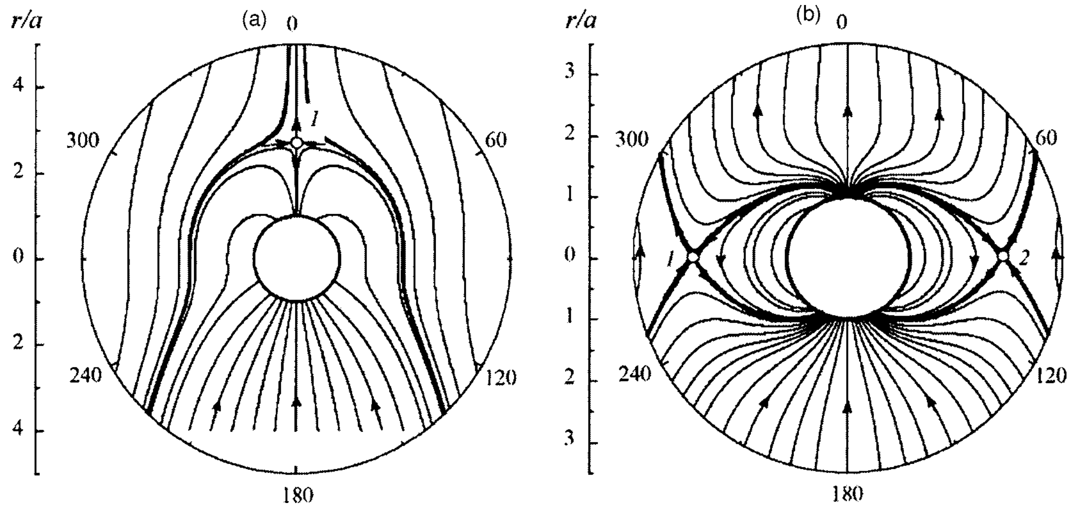

FIG. 1. Pattern of field lines constructed for a particle with $q=0.3$ and $n^2=-2+0.2i$ from the formulas of the dipole approximation (a) and from the exact formulas of Mie theory (b).

(the Poynting vector). The time-averaged Poynting vector is determined by

$$\langle \mathbf{S} \rangle = \frac{c}{4\pi} \mathrm{Re}(\mathbf{E} \times \mathbf{H}^*), \tag{19}$$

where the asterisk denotes the operation of complex conjugation. The fields around a particle are sums of the incident and scattering fields—for example, the electric field $\mathbf{E}=\mathbf{E}^{(i)}+\mathbf{E}^{(s)}$. In the case of Rayleigh scattering, it is sufficient to take into account only one dipole term in the expansion of the fields in spherical harmonics for scattered radiation fields $\mathbf{E}^{(s)}$ and $\mathbf{H}^{(s)}$. If the incident plane wave propagates in direction z, while the electric field vector in it is directed along the x axis, the field lines always remain in this plane,[23] while the corresponding equation for the field lines in spherical coordinates has the form

$$\frac{dr}{d\theta} = r \frac{\langle S \rangle_r}{\langle S \rangle_\theta}. \tag{20}$$

Here $\langle S \rangle_r$ and $\langle S \rangle_\theta$ are the corresponding spherical components of the Poynting vector, while $\langle S \rangle_\varphi$ equals zero in the case under consideration. Substituting the fields found from Mie theory into Eq. (20), it is possible after an number of transformations to obtain the corresponding equation in explicit form:[23]

$$\frac{d\rho}{d\theta} = -\rho c \tan \theta \frac{F(\rho, \theta)}{G(\rho, \theta)}, \tag{21}$$

where functions F and G are determined by

$$F = \rho^3 + (q^2\rho^2 \cos \theta + q^2\rho^2 - 1)(K_r \cos \xi + K_i \sin \xi) + (q\rho \cos \theta + q\rho)(K_r \sin \xi - K_i \cos \xi),$$

$$G = \rho^3 + (q^2\rho^2 \cos \theta + 2)(K_r \cos \xi + K_i \sin \xi) + (q\rho \cos \theta - 2q\rho)(K_r \sin \xi - K_i \cos \xi). \tag{22}$$

In these formulas, $K=K_r+iK_i=(n^2-1)/(n^2+2)$, $\xi=q\rho(\cos \theta-1)$, and $\rho=r/a$.

The corresponding fields can have singular points $\rho=\rho_i$, $\theta=\theta_i$, lying at the intersections of the null isolines $F(\rho,\theta)=0$ and $G(\rho,\theta)=0$. The character of these singular points is determined by the coefficients of the linear expansions of the corresponding functions. Since the medium around the particle is nonabsorbing (vacuum), div $\mathbf{S}=0$ in this medium (see §80 in Ref. 12). According to the common assumptions of the theory of nonlinear vibrations,[24] singular points that satisfy this condition on a plane can be only centers or saddles. In an absorbing medium, however, div $\mathbf{S}<0$, and the corresponding singular points can be nodes or foci.

The Poynting-vector lines constructed from Eqs. (21) and (22) are shown in Fig. 1a. It can be seen from the figure that there is a single saddle point *1* in the case of Rayleigh scattering at a plane, while the field lines themselves enter into the particle from all directions.[23] If one is not limited to the dipole approximation and the field lines are constructed from the exact formula of Mie theory, the field pattern shown in Fig. 1b is obtained.[18] In the latter case, there are two saddle points *1* and *2*, while the energy flows into and out of the particle.

The cause of the obvious discrepancy of the two figures is associated with the fact that Eq. (22) neglects effects caused by radiative emission of a plasmon. As was shown in Ref. 18, a Rayleigh-type pattern appears only for sufficiently large dissipation (for a particle with $q=0.3$, when Im $n^2>0.58$), whereas the pattern shown in Fig. 1b occurs in the absence of dissipation. Wang *et al.*[18] called the corresponding field pattern *Tribelskiĭ ears*. As was shown in Ref. 18, the field patterns in Figs. 1a and 1b correspond to the limiting cases of large and small dissipation. The phase portrait of the field lines can undergo several transformations as the absorption increases, as a result of which structures of the type of optical vortices appear, along with structures with stable and unstable nodes. This does not contradict the condition div $\mathbf{S}=0$, which must be satisfied in three-dimensional space, where the corresponding singular points can be saddle-nodes and saddle-foci.[18] Figure 2 shows examples of patterns of

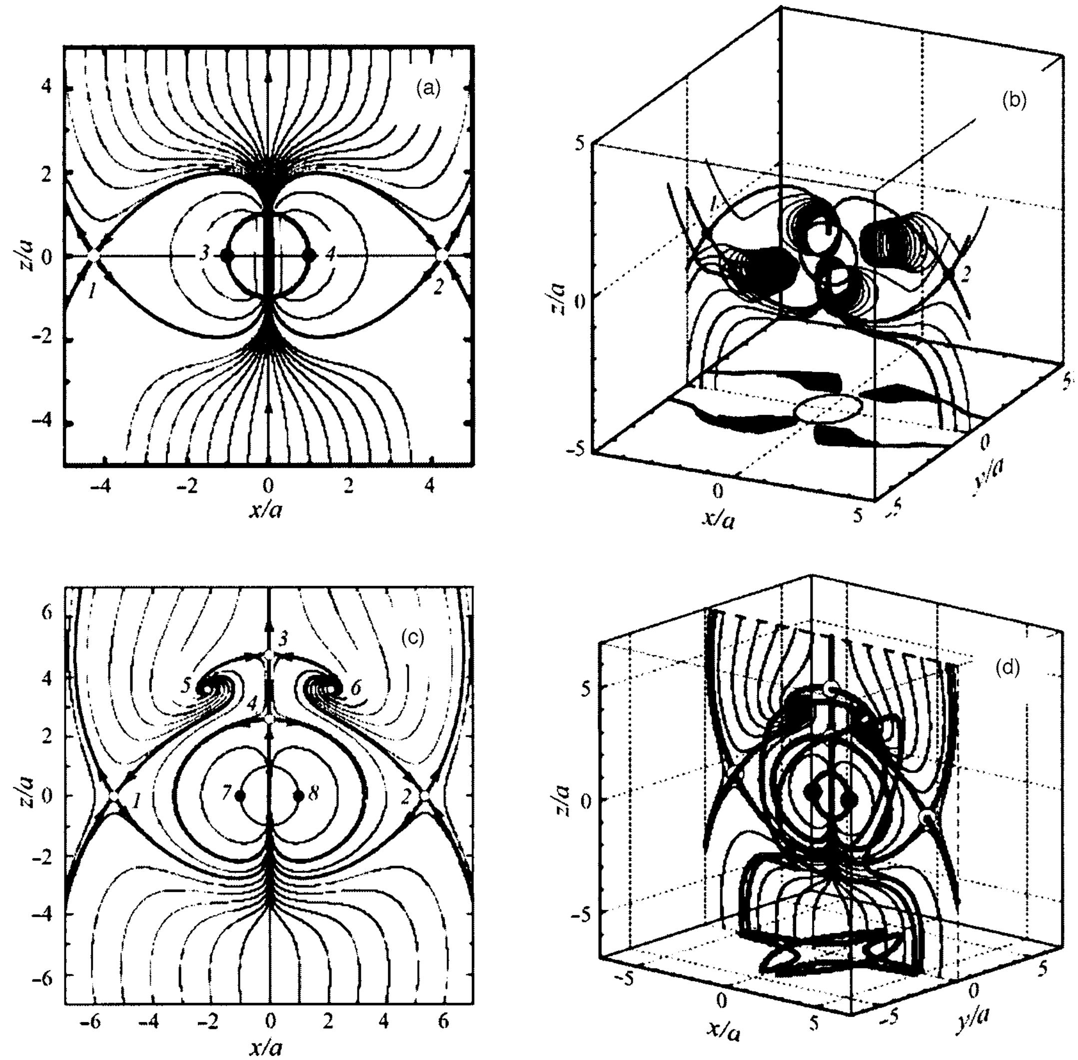

FIG. 2. Two-dimensional and three-dimensional patterns of field lines computed for a particle with $q=0.3$ in the absence of dissipation. The values of the parameter are $n^2=-2.1$ in (a) and (b) and $n^2=-2.17$ in (c) and (d). In (a), points *1* and *2* are saddle points, and points *3* and *4* are *centers*. In (c), points, *1*, *2*, *3*, and *4* are saddle points, points *5* and *6* are *foci*, and points *7* and *8* are *centers*.

field lines for a particle with $q=0.3$ in the absence of dissipation.

It can be seen from the figures that the Tribelskiĭ ears are regions with closed Poynting-vector lines; i.e., a cyclic energy flow takes place around the singular points on the surface of the particle (points *3* and *4* in Fig. 2a and points *7* and *8* in Fig. 2c). These points are not ordinary centers in the sense of the theory of vibrations,[24] since they are not points of intersection of null-isoclinic lines. We recall that the Poynting-vector lines inside and outside the particle are determined from different equations. The field lines themselves are continuous in this case, but they experience "refraction" at the boundaries because of the boundary conditions for the normal component of the electric field.

These *center-type* singular points, around which energy "rotates," correspond to the maxima of the Pointing vector $|\mathbf{S}|$ where $S_r=0$. When a small dissipation is switched on, these points transform into *foci*, as a result of which the divergence of the field vector becomes negative at the corresponding points on the boundary, $\operatorname{div}\mathbf{S}<0$. These points also are not ordinary foci in the classical theory of vibrations. Reference 19 proposed to call the structure of the energy flow around such energy sinks an *optical whirlpool*. As the dissipation increases, the optical whirlpool transforms into a node and its center escapes from the surface into the depth of the particle. The corresponding singular points in this case are nodes in their usual meaning in the theory of vibrations.

A localized plasmon, according to the studies of Refs. 18 and 19, is thus associated with the rotational structure of the energy flow around the singular points at the boundary of the particle. This structure is stable and weakly depends on the geometry. As was shown in Ref. 25, optical whirlpools, for example, exist for spheroidal particles. Analogous structures were found in Ref. 25 for thin metallic cylinders. Figure 3 shows such a structure for polarized radiation whose **E** vector is parallel to the y axis, while the radiation is incident on the particle from the right.

Besides circular energy flows around singular points at the center of localized plasmons, other singularities can appear as the parameters q, $\operatorname{Re} n^2$, and dissipation $\operatorname{Im} n^2$ vary

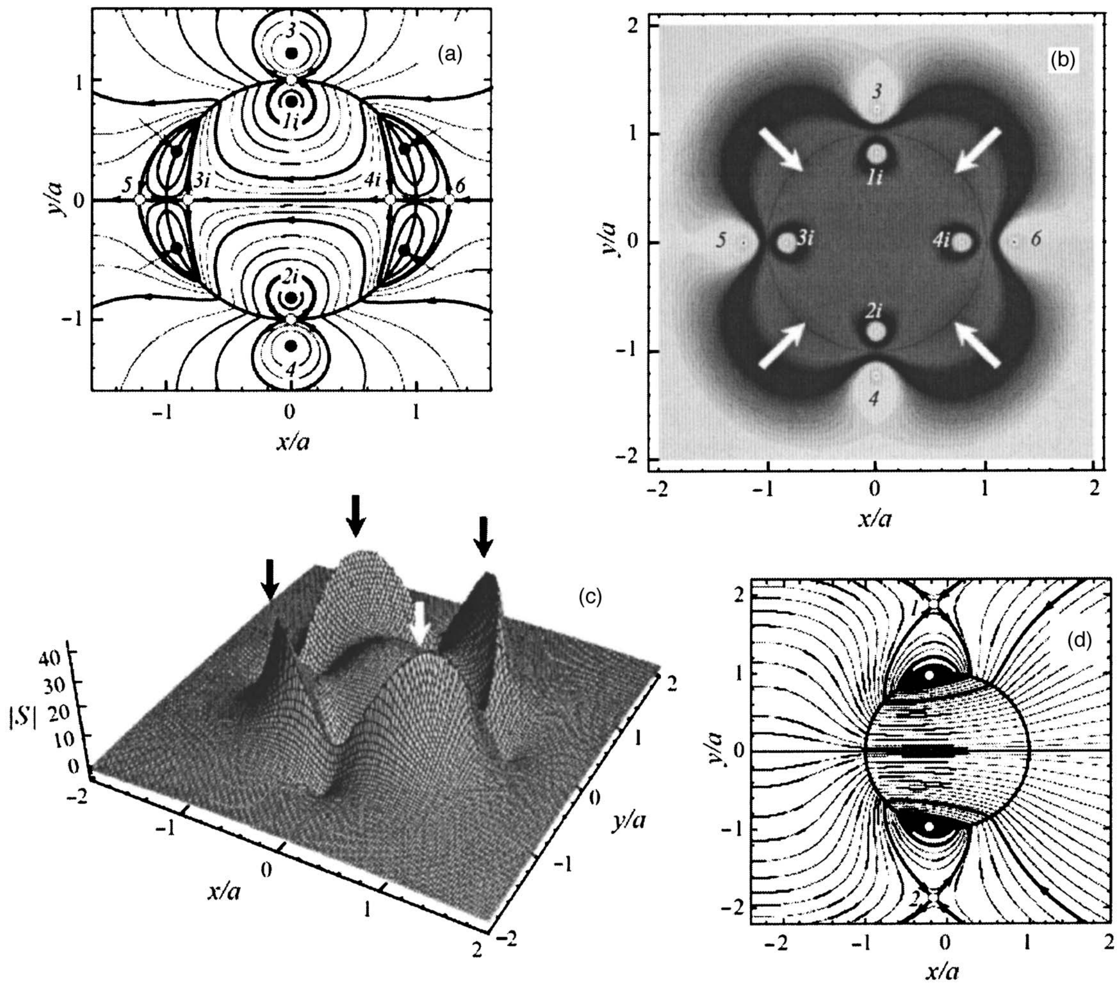

FIG. 3. Poynting-vector lines for light scattering at a thin metallic cylinder: $q=0.1$ and $n^2=-1$ (a) and the corresponding contour graph (b) and three-dimensional graph (c) of the quantity $|\mathbf{S}|$. The arrows denote the four singular points (*centers*) around which the energy flows circulate. These points correspond to local maxima of the Poynting vector, as is seen on the three-dimensional graph (c). When dissipation is switched on, the *centers* transform into *foci*, which are optical whirlpools; these optical whirlpools in a medium with $q=0.1$ and $n^2=-1+0.1i$ are shown in graph (d).

on the phase plane of the energy flow—in particular optical vortices, which are flows that appear around singular points of the saddle-focus type.[18]

Optical vortices can even appear when light is scattered at a particle that does not produce dissipation, as shown in Fig. 2c (the corresponding singular points are *5* and *6*). The energy deflected from these points in the xz plane is precisely compensated by the supply of energy from energy flows directed toward each other in the perpendicular direction. The corresponding field lines are shown in the three-dimensional Fig. 2d. As a result, the condition for the three-dimensional Poynting vector $\mathrm{div}\,\mathbf{S}=0$ is satisfied in the entire region outside the particle.

Figure 2b shows a three-dimensional pattern of force lines for an energy flow deflected from regions with circulation of the Poynting vector on the xz plane. The field lines represent helicons nested in each other, deflecting energy to infinity (the directions in which the energy is deflected can be easily seen on the projection of the corresponding flows onto the xz plane).

The near-field pattern of the energy flow close to a particle with a plasmon resonance thus radically differs from the Rayleigh pattern. These differences can even be observed in the far field, for example, in the form of reverse hierarchy of the resonances;[17,22] however, this requires media with a very small dissipation. The structure of the Poynting-vector lines varies in the near-field pattern even for media with large dissipation.[18] Since the structure of the energy flow can be reconstructed as the radiation frequency varies, this provides a surprising possibility for manipulating energy flows at small scales (switching on and off optical resonances and varying the number of singular points as well as the directions of energy flow).

The authors are grateful to S. I. Anisimov and L. P. Pitaevskiĭ for discussing a number of questions relating to this work. Part of these investigations were carried out with the support of the Russian Foundation for Basic Research (Grant 04-02-16972).

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[1)]In *The Star Diaries of Ijon Tichy*, Stanislav Lem described an astonishing invention of Ijon's—a light bulb that produces darkness when it is turned on. Ijon Tichy apparently filled the light bulb with a gas of Rayleigh particles with low dissipation, in which the plasmon frequencies covered the entire visible spectrum. When it was "turned on," curtains were simply drawn back that allowed the particles to efficiently scatter the surrounding light, as a result of which Ijon Tichy found himself in deep shadow. The attentive reader had of course grasped that Ijon Tichy's second invention, noninflammable matches for children, is also based on the use of plasmon resonance in weakly dissipative media. Since the adhesion coefficient, according to the Lifshitz theory, is determined by $[(n^2-1)/(n^2+1)]^2$ [see Eq. (82.6) in Ref. 16], for sufficiently high adhesion, when n^2 is close to unity, a child simply does not have the strength to strike a match.

2)For purely real n^2, the values of $\mathcal{R}_l$ and $\mathcal{J}_l$ in Eq. (5) are proportional to n^l multiplied by a purely real quantity, and as a result $\mathrm{Re}\, a_l = \mathcal{R}_l^2/(\mathcal{R}_l^2+\mathcal{J}_l^2)$. Analogously, $|a_l|^2 = \mathcal{R}_l^2/(\mathcal{R}_l^2+\mathcal{J}_l^2)$; i.e., in the absence of dissipation, it is always true that $Q_{\text{sca}} \equiv Q_{\text{ext}}$. However, if the approximation given in Eq. (9) is used, a paradoxical result is obtained: whereas, for Q_{sca}, this approximation gives Eq. (10), an identical zero is obtained for Q_{ext} (which at least cannot be less than Q_{sca}). The equality $Q_{\text{sca}} = Q_{\text{ext}}$ is restored only if, in Eq. (3), for Q_{ext} one substitutes amplitude a_1, taking into account the first orders in the small ratio $\mathcal{R}_l/\mathcal{J}_l$. This fact now brings to mind that neglecting small $\mathcal{R}_l/\mathcal{J}_l$ can lead to an erroneous result.

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