Resonant Behavior of Dielectric Objects (Electrostatic Resonances)

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Resonant behavior of dielectric objects occurs at certain frequencies for which the object permittivity is negative and the free-space wavelength is large in comparison with the object dimensions. Unique physical features of these resonances are studied and a novel technique for the calculation of resonance values of permittivity, and hence resonance frequencies, is proposed. Scale invariance of resonance frequencies, unusually strong orthogonality properties of resonance modes, and a two-dimensional phenomenon of "twin" spectra are reported. The paper concludes with brief discussions of optical controllability of these resonances in semiconductor nanoparticles and a plausible, electrostatic resonance based, mechanism for nucleation and formation of ball lightning.

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Small dielectric objects can exhibit resonance behavior at certain frequencies for which the object permittivity is negative and the free-space wavelength is large in comparison with object dimensions. The latter condition clearly suggests that these resonances are electrostatic in nature. They appear at specific negative values of dielectric permittivity for which source-free electrostatic fields may exist. This is, in essence, the physical mechanism of these resonances. For nanoscale metallic objects, these resonances occur in the optical frequency range and they result in powerful localized sources of light that are useful in scanning near-field optical microscopy [1,2], nanolithography [3], and in biosensor applications [4,5]. It is also believed [6] that strong local electromagnetic fields associated with these resonances may play an important role in surface enhanced Raman scattering [7–9]. Currently, such resonances are found experimentally (or numerically) by probing dielectric objects of complex shapes with radiation of various frequencies [10–13]. General physical properties of these resonances have not been studied and there has not existed any technique for direct calculation of negative values of dielectric permittivity, and the corresponding frequencies of electromagnetic radiation, for which resonances occur. The purpose of this paper is to develop such a technique as well as to study unique physical features of electrostatic resonances. It is demonstrated that the resonance values of permittivity, and hence the resonance frequencies, can be directly (i.e., without laborious probing) found by computing the eigenvalues of a specific boundary integral equation. This approach reveals the unique physical property of electrostatic resonances: resonance frequencies depend on dielectric object shapes, but they are scale invariant with respect to geometric dimensions. Unusually strong orthogonality properties of resonance modes are obtained. These orthogonality properties are physically important for the selection of resonance modes that can be coupled to incident electromagnetic radiation. It is found that in the case of nanowires (i.e., two-dimensional particles) the physical phenomenon of twin spectra occurs where resonance values of relative dielectric permittivity are arranged in pairs of reciprocal negative numbers. *A priori* estimates of upper and lower bounds for resonance frequencies are obtained for convex dielectric objects. These estimates effectively narrow the frequency range of possible resonances. The paper is concluded with a brief discussion of a plausible, electrostatic resonance based, mechanism for nucleation and formation of ball lightning.

To start the discussion, consider a dielectric object of arbitrary shape with permittivity ε (Fig. 1). We are interested in such negative values of ε for which a source-free electrostatic field may exist. This source-free field is curl and divergence free inside (V^+) and outside (V^-) of the dielectric object; its electric potential is continuous across the boundary S of the object, while the normal components of the electric field satisfy the boundary condition:

$$\varepsilon E_n^+ = \varepsilon_0 E_n^- \text{ on } S. \tag{1}$$

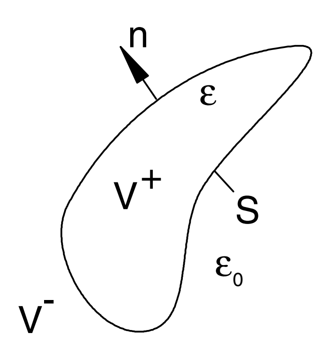

FIG. 1. The dielectric region V^+ bounded by the surface S.

The electric potential of this source-free field can be represented as an electric potential of a single layer of electric charges σ distributed over S:

$$\varphi(Q) = \frac{1}{4\pi\varepsilon_0}\int_S \frac{\sigma(M)}{r_{MQ}} dS_M. \tag{2}$$

In other words, a single layer of electric charges σ on S creates the same electric field in the free space as the source-free electric field that may exist in the presence of the dielectric object. It is apparent that the electric field of surface charges σ is curl and divergence free in V^+ and V^- and its potential is continuous across S. To satisfy the boundary condition (1), we recall that the normal components of the electric field of a single layer potential are given by the formulas [14,15]

$$E_n^{\pm}(Q) = \mp\frac{\sigma(Q)}{2\varepsilon_0} + \frac{1}{4\pi\varepsilon_0}\int_S \sigma(M)\frac{\mathbf{r}_{MQ}\cdot\mathbf{n}_Q}{r_{MQ}^3} dS_M. \tag{3}$$

By substituting (3) into the boundary condition (1), we arrive at the homogeneous boundary integral equation

$$\sigma(Q) = \frac{\lambda}{2\pi}\int_S \sigma(M)\frac{\mathbf{r}_{MQ}\cdot\mathbf{n}_Q}{r_{MQ}^3} dS_M, \tag{4}$$

where

$$\lambda = \frac{\varepsilon - \varepsilon_0}{\varepsilon + \varepsilon_0}. \tag{5}$$

Thus, source-free electric fields may exist only for such values of permittivity ε that integral equation (4) has nonzero solutions. In other words, in order to find the resonance values of ε (and the corresponding resonance frequencies) as well as resonance electrostatic modes, the eigenvalues and eigenfunctions of the integral equation (4) must be found.

Since the integral operator in (4) is compact, the spectrum is discrete. It can be shown that the spectrum has the following interesting properties: all eigenvalues are real, $\lambda = 1$ is an eigenvalue, and for all other eigenvalues $|\lambda| > 1$ [16,17]. The eigenvalue $\lambda = 1$ corresponds to the case of $\varepsilon \to \infty$ and the respective eigenfunction $\sigma(M)$ is the distribution of surface electric charges over the surface S of conductor V^+. This eigenvalue is irrelevant to our discussions. All other eigenvalues correspond to source-free (resonance) configurations of electrostatic fields and, according to (5), these configurations may exist (as expected) only for negative values of ε. After these negative resonance values of ε are found through the solution of integral equation (4), the appropriate frequency dependent permittivity $\varepsilon(\omega)$ can be employed to find the resonance frequencies.

It is apparent that the mathematical structure of integral equation (4) is invariant with respect to the scaling of S, i.e., the scaling of the dimensions of the dielectric object. This leads to the unique property of electrostatic resonances: resonance frequencies depend on object shape but they are scale invariant with respect to dielectric object dimensions, provided that they remain appreciably smaller than the free-space wavelength.

The integral operator in Eq. (4) is not Hermitian (not self-adjoint), because the kernel of this equation is not symmetric. For this reason, the eigenfunctions $\sigma_i(M)$ and $\sigma_k(M)$ corresponding to different eigenvalues λ_i and λ_k are not orthogonal on S. Nevertheless, it can be shown that the electric fields $\mathbf{E}_i$ and $\mathbf{E}_k$ corresponding to eigenfunctions σ_i and σ_k satisfy

$$\int_{V^\pm} \mathbf{E}_i \cdot \mathbf{E}_k dV = 0. \tag{6}$$

The peculiar feature of the above orthogonality conditions is that they hold separately in regions V^+ and V^-.

These orthogonality conditions can be useful in the analysis of the coupling of a specific resonance mode to incident electromagnetic fields. For instance, for a sphere and for ellipsoids there are resonance modes with uniform electric fields in V^+ (see below). This means, according to the orthogonality condition, that only these "uniform" resonance modes will be excited by uniform (within V^+) incident radiation. The condition of uniformity within V^+ of the incident radiation is to some extent natural due to the smallness of object dimensions in comparison with the free-space wavelength of the incident radiation. For dielectric objects of complex shapes, many resonance modes with appreciable average values of electric field components over V^+ may exist. All such modes will be well coupled to the uniform incident radiation and can be excited by such incident fields at the respective resonance frequencies.

It can be shown that for two-dimensional objects (i.e., nanowires) an interesting phenomenon of twin spectra occurs where the set of eigenvalues consists of pairs of real numbers symmetric with respect to the origin. According to (5), this means that the set of resonance values of the relative dielectric permittivity consists of pairs of reciprocal negative numbers. This twin spectrum phenomenon occurs for two-dimensional objects of any cross-sectional shapes.

For convex S, the following estimate for the eigenvalues λ can be derived:

$$|\lambda| > c = \frac{1}{1 - \frac{A}{4\pi R d}}, \tag{7}$$

where A is the area of S, R is the maximum radius of the curvature of S, and d is the diameter of V^+. By using the last inequality and (5), the following upper and lower bounds for possible resonance values of permittivity ε can be obtained:

$$\frac{1+c}{1-c} < \frac{\varepsilon}{\varepsilon_0} < \frac{1-c}{1+c}. \tag{8}$$

For the common plasma permittivity $\varepsilon/\varepsilon_0 = 1 - \omega_p^2/\omega^2$, the last formula leads to the following upper and lower bounds for resonance frequencies:

$$\frac{c-1}{2c} < \frac{\omega^2}{\omega_p^2} < \frac{c+1}{2c}, \tag{9}$$

which suggests that the bandwidth for resonance frequencies is smaller than $\omega_p/\sqrt{c} = \omega_p\sqrt{1 - A/2Rd}$.

Consider two examples such as the following.

First, in the case of a unit dielectric sphere, the kernel of integral equation (4) is equal to $1/2r_{MQ}$. By using this fact and the spherical harmonic addition theorem, it is easy to demonstrate that the spherical harmonics $Y_{\ell m}(\theta, \phi)$ are the eigenfunctions corresponding to the eigenvalue $\lambda_\ell = 2\ell + 1$. According to (5), the corresponding resonance values of permittivity are $\varepsilon_\ell = -\varepsilon_0(1 + 1/\ell)$ $(\ell \geq 1)$. Because of the scale invariance, the same ε_ℓ are the resonance permittivity values for a dielectric sphere of arbitrary radius. The three lowest electrostatic modes corresponding to $\varepsilon_1 = -2\varepsilon_0$ are uniform in V^+ and only these modes can be excited by uniform (within V^+) incident radiation.

Second, consider a dielectric ellipsoid. In this case, the resonance permittivity values corresponding to uniform source-free electric fields in V^+ can be found without solving integral equation (4). Indeed, the source-free uniform electric fields $\mathbf{E}^+$ inside the ellipsoid must satisfy the homogeneous equations:

$$\frac{\varepsilon}{\varepsilon_0}\mathsf{N}\mathbf{E}^+ + (\mathsf{I} - \mathsf{N})\mathbf{E}^+ = 0, \tag{10}$$

where N is (for the appropriate choice of axes) a diagonal matrix of depolarizing coefficients, while I is the identity matrix. Nonzero solutions of Eq. (10) exist for such values of ε that the diagonal matrix $\mathsf{I} + (\varepsilon/\varepsilon_0 - 1)\mathsf{N}$ is singular. It follows that spatially uniform (inside the ellipsoid) electrostatic resonances may exist only for the special values of permittivity

$$\varepsilon_i = \varepsilon_0(1 - 1/N_i), \tag{11}$$

where N_i are diagonal entries of N. It follows from the orthogonality conditions (6) that for all other resonance electrostatic modes the mean values of electric field components in V^+ are equal to zero. For this reason, all other resonance modes cannot be excited by spatially uniform incident radiation. It is apparent from (11), that for any negative value of ε an appropriate ellipsoid can be found that will "resonate" for this ε. In the case of the plasma $\varepsilon = \varepsilon_0(1 - \omega_p^2/\omega^2)$, the last assertion means that any frequency $\omega < \omega_p$ can be a resonance frequency for an appropriate ellipsoid. Finally, from the above dispersion relation and the condition $\sum_i N_i = 1$ it follows that $\sum_i \omega_i^2 = \omega_p^2$, where ω_i are the resonance frequencies "along" the main axes.

For dielectric objects of complex shapes the resonance frequencies and resonance modes can be found through numerical solution of integral equation (4). If the boundary S of the dielectric object is irregular, then $\sigma(M)$ may have singularities at the corners and the edges of S. In this situation, it may be more convenient to solve the integral equation

$$\tau(Q) = \frac{\lambda}{2\pi}\int_S \tau(M)\frac{\mathbf{r}_{QM}\cdot\mathbf{n}_M}{r_{QM}^3}dS_M \tag{12}$$

that is adjoint to Eq. (4) and has the same spectrum. Equation (12) has a simple physical meaning. Solutions $\tau(M)$ of the integral equation (12) signify dipole densities, i.e., densities of a double layer of electric charges distributed over S that create the same electric displacement field ($\mathbf{D}$) as the source-free electric displacement field that may exist in the presence of dielectric objects with negative ε.

By solving integral equation (4) [or Eq. (12)], the electric fields of electrostatic modes can be found. By using these fields, multipoles of dielectric objects can be computed and then can be used to evaluate radiation losses (and quality factors) associated with electrostatic resonances. However, the *near field* is given by the appropriate integral over the equivalent charge distribution σ, which will be quite different from a superposition of multipole fields in all cases except for a sphere. Corrections to electrostatic resonance modes due to radiation can be found by using series expansions of the solutions to time harmonic Maxwell equations with respect to the small ratio of the object diameter to the free-space wavelength. It is apparent that, unlike electrostatic resonance modes, these radiative corrections are not scale invariant.

Our discussion can be easily extended to the analysis of electrostatic resonances of several dielectric objects located in proximity to one another. In this case, S in integral equation (4) must be construed as the union of boundaries of all dielectric objects and σ as being defined on this union. The spectral properties of integral equation (4) and the orthogonality conditions are the same as in the case of a single dielectric object.

We next present some illustrative calculations in two dimensions, i.e., for infinite cylinders. The two-dimensional version of (4) is

$$\sigma(Q) = \frac{\lambda}{\pi}\int_S \sigma(M)\frac{\mathbf{r}_{MQ}\cdot\mathbf{n}_Q}{r_{MQ}^2}dS_M, \tag{13}$$

where S is now the boundary curve with line element dS, and the potential is the logarithmic potential created by the line charge with density σ. We first investigated ellipses (infinite elliptic cylinders), for which the spectrum can be found analytically. An ellipse with semimajor axis a and semiminor axis b has spectrum $\lambda_0 = 1$ and

$$\lambda_n^\pm = \pm\left[\frac{a+b}{a-b}\right]^n. \tag{14}$$

The eigenvalues $\lambda_1^\pm$ correspond to a uniform field in V^+ and are related to the depolarization coefficients

$$N_a = \frac{b}{a+b}, \qquad N_b = \frac{a}{a+b}. \tag{15}$$

The dipole moments associated with all other modes are

TABLE I. Spectrum of an ellipse with 5:1 aspect ratio.

Numerical	Exact	Numerical	Exact
1.0010	1.0000		
−1.4992	−1.5000	1.5037	1.5000
−2.2472	−2.2500	2.2573	2.2500
−3.3670	−3.3750	3.3898	3.3750
−5.0419	−5.0625	5.0932	5.0625
−7.5437	−7.5938	7.6592	7.5938

zero. Table I shows numerical results for an ellipse with $a/b = 5$.

We also studied the spectrum of an equilateral triangle with rounded corners. In this case, the spectrum is not known *a priori*; however, certain qualitative features of this spectrum can be predicted on the basis of symmetry. Indeed, the above triangle is invariant with respect to the transformations of the group C_{3v} [18]. This group has three inequivalent irreducible representations: two of dimension one and one of dimension two. This fact implies that the spectrum may consist only of simple and twofold degenerate eigenvalues and that the dipole moments of resonance modes corresponding to simple eigenvalues are equal to zero. Table II shows our numerical results, which are consistent with these qualitative features of the spectrum. It is clear on the symmetry grounds that the same spectral features are also valid for a three-dimensional prism with the same cross section. It is also clear that similar qualitative spectral features can be predicted for other symmetric three-dimensional dielectric objects by using irreducible representations of their symmetry groups.

It is worthwhile to point out that electrostatic resonances in semiconductor nanoparticles are of special interest because they can be controlled through optical manipulation of the carrier densities. This optical controllability can be utilized for the development of nanoscale light switches and all-optical nanotransistors.

TABLE II. Numerical results for the rounded triangle.

Eigenvalue (λ)	d_x	d_y
1.0015	0.0000	0.0000
−2.4459	−0.1681	−0.3028
−2.4459	−0.3094	0.1556
2.4696	−0.0472	−0.6230
2.4696	−0.6244	0.0200
−4.3263	−0.0000	−0.0000
4.3809	0.0000	−0.0000
−13.3646	−0.0421	0.0656
−13.3646	0.0628	0.0461
13.5374	−0.0267	0.0808
13.5374	0.0149	−0.0838

We conclude this paper with a brief discussion of a plausible explanation of the phenomenon of ball lightning based on electrostatic resonances. This enigmatic natural phenomenon usually occurs after a lightning strike that may lead to plasma formation and serve as a source of considerable electromagnetic radiation [19,20]. If the frequency spectrum of this radiation is such that the dielectric permittivity of the formed plasma is negative, then electrostatic resonances may occur. The nucleation of electrostatic resonances and the spatial growth of resonance regions may be facilitated by the scale invariance of the resonance frequencies. Electrostatic resonances may produce a considerable localized accumulation of electromagnetic energy that may visually manifest itself as ball lightning.

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