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Plasmon oscillations in a linear cluster of spherical nanoparticles

I.V. Zabkov, V.V. Klimov, I.V. Treshin, O.A. Glazov

Abstract. **The eigenfrequencies and eigenfunctions of plasmon oscillations in an infinite linear cluster of spherical nanoparticles are found analytically within the framework of the quasi-static approach and numerically within the framework of the finite element method. It is shown that the spectrum of plasmon oscillations is complex and contains previously unknown, highly localised modes in a high-frequency region of the spectrum. The results obtained can be used in designing plasmon nanowaveguides and nanolasers.**

Keywords: *nanoparticles, nanowaveguide, plasmon modes, nanoplasmonics*.

1. Introduction

Currently, thanks to the success of nanotechnology, nanoplasmonics (a field of nanophotonics), which investigates the collective oscillations of conduction electrons in metal particles, is being actively developed [1]. One of the important tasks of nanoplasmonics is to study the optical properties of linear and other clusters, resulting, in particular, from the desire to use arrays of plasmon nanoparticles as waveguides. This may allow the integration of advanced silicon electronics with high-performance optical plasmon waveguides [2 – 8].

The exact solution to the scattering problem of an electromagnetic wave on a spherical particle was found more than 100 years ago by Mie [9] and independently by Debye [10]. In 1979, Bergman [11], solving the quasi-static problem aimed at finding the effective dielectric constant of a two-component medium with spherical inclusions, proposed to expand eigenfunctions – and in fact, the potential inside the particles – in spherical harmonics with the centre in each sphere. A similar approach was used by Gérardy [12], who considered the problem of finding the absorption spectrum of the cluster consisting of several spheres of arbitrary size. An important result of papers [11, 12] was the calculation of the coefficients that describe the interaction between the two multipole potential distributions located at different points. It may be noted that similar coefficients were calculated by Claro [13].

A good understanding of the physical processes that occur when light interacts with the clusters can be achieved by using a model in which each particle is approximated by a point electric dipole, while quadrupole and higher moments are ignored. At the same time Claro [14], studying a cluster consisting of two, three and an infinite number of particles showed that the dipole model works well as long as the distance between the centres of the particles are three times larger than their radius. This model is used to calculate the dispersion curves of plasmon modes in a linear cluster in the quasi-static case [2] and in the case of when the retardation is taken into account [15], as well as to calculate the energy transfer by a linear chain of nanoparticles in the quasi-static approximation [16]. Weber and Ford [15] also considered the effect of absorption in the particles on the dispersion curves of propagating plasmons. Koenderink and Polman [17] showed that at the point of intersection of the dispersion curves of plasmons with the dispersion curve of photons in free space we can observe not only a serious discrepancy between the results obtained by taking into account the retardation in the quasi-static case, but also a break in the dispersion curves.

The question about the impact of higher-order multipoles on the optical properties of the cluster rises in many works. For example, in the quasi-static case the multipole interaction was considered in [13, 14, 18]. The authors of papers [19 – 21] solved the quasi-static problem for two spheres in the bispherical coordinates, each term of the expansion of the potential in these coordinates taking into account already an infinite number of ordinary multipoles. In paper [22] the problem for two cylinders was exactly solved in the bipolar coordinates.

The analytical solution to the problem of interaction of light with clusters of spherical particles with allowance for the retardation effects was first obtained by Gérardy [23] while calculating their absorption cross section. In this case the electromagnetic field was expanded in vector spherical harmonics. Later this problem was considered by many authors, including Mackowski [24] and Xu [25]. The distribution of plasmon oscillations along the cluster was first studied in [26]. Chern et al. [27] investigated the linear cluster consisting of several spherical particles (up to twelve) and placed in an external field. Pinchuk [28] studied the

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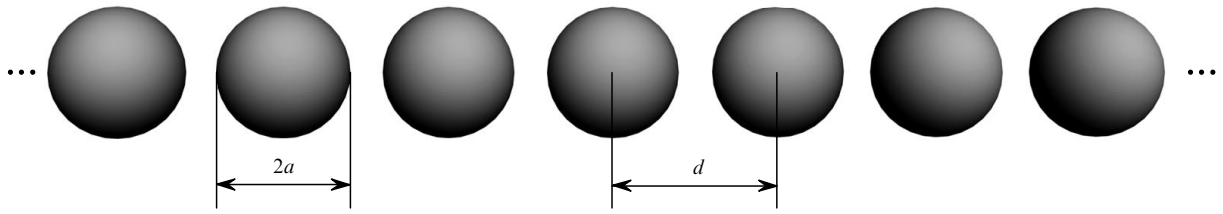

Figure 1. Geometry of the problem.

dependence of the intensity of light scattering by a linear cluster, consisting of ten nanospheres, on the angle of incidence of the external plane wave. Khlebtsov et al. [29] compared the dipole and multipole solutions to the problem of scattering of light by a dimer. To calculate the optical properties of the clusters, numerical methods are actively used, including the finite difference time domain (FDTD) method [3], finite element method (FEM) [30] and the discrete dipole approximation (DDA) [31, 32]. Andersen et al. [32] numerically investigated the optical properties of different configurations of spherical particles and compared the results obtained by the methods used in [23, 25], as well as by the discrete dipole approximation with one or more dipoles in the particle. The influence of the substrate was considered in [33–35]. Demidenko et al. [36] studied the optical properties of the cluster of magnetic-plasmonic particles on an insulating substrate.

However, up to date the structure of the plasmon eigenoscillations and the corresponding eigenfrequencies of a linear cluster of spherical nanoparticles have been studied neither in the quasi-static approximation, nor in case of a retardation problem. Understanding the structure of the eigenfunctions, i.e., the potential and field distributions, as well as the dependence of plasmon frequencies on the cluster geometry is important for describing the behaviour of the cluster in an external field. In this case, along with the external plane waves, it is possible to investigate the interaction of a cluster with localised radiation of atoms and molecules. Eigenoscillations (in the absence of the external field) of clusters of nanoparticles can be found from the solution of homogeneous Maxwell equations. Fields, which arise when placing the cluster in an arbitrary external field, can be found by their series expansion in the eigenfunctions [1].

The aim of this work is to find and analyse of eigenoscillations of a linear cluster of spherical metal nanoparticles, i.e., the dispersion curves of emerging plasmons and spatial potential distributions corresponding to different plasmon modes.

The geometry of the problem is shown in Fig. 1. We will use the formulation of the problem, similar to that used in [11]. To calculate the spatial distribution of the potentials we have also performed simulations by the finite element method whose results were in good agreement with the results obtained analytically.

2. Analytic solution to the problem of eigenoscillations of a linear cluster of spherical nanoparticles

Consider first an arbitrary cluster consisting of spherical nanoparticles. Let it be located in an infinite homogeneous and isotropic space with the dielectric constant ε_h. The sphere with the number α has the radius a_α and dielectric constant ε.

To find in the quasi-static approximation plasmon eigenmodes of nanoparticles of any shape and of their clusters, it is needed to solve the homogeneous – without sources – problem of potential theory, i.e., the Laplace equation with the requirement of continuity of the potential and normal component of electric induction at the boundary of each sphere and with the condition of the potential vanishing at infinity.

For a sphere of radius a_0, this problem has a well-known solution [1], and the eigenfunctions ψ_{lm} and eigenvalues of the dielectric constant ε_l have the form

$$\psi_{lm}(\boldsymbol{r}) = \begin{cases} r^l Y_l^m(\theta,\varphi), & r \leqslant a_0, \\ \dfrac{a_0^{2l+1}}{r^{l+1}} Y_l^m(\theta,\varphi), & r > a_0, \end{cases} \qquad \frac{\varepsilon_l}{\varepsilon_h} = -\frac{l+1}{l},$$

$$l = 1, 2, \ldots, \; m = -l \ldots l, \tag{1}$$

$$Y_l^m(\theta,\varphi) = (-1)^m \left[\frac{2l+1}{4\pi}\frac{(l-m)!}{(l+m)!}\right]^{1/2} P_l^m(\cos\theta)\exp(\mathrm{i}m\varphi),$$

where $Y_l^m(\theta,\varphi)$ are the spherical harmonics; $P_l^m(\cos\theta)$ are the associated Legendre polynomials [37]; r, θ, φ are the spherical coordinates associated with the centre of the sphere. The case $l = 0$ corresponds to the trivial case of a static charge distributed over the surface of the sphere. A similar solution holds for the linear cluster, but we do not take it into account.

To describe the field near an arbitrary particle of the cluster, say a particle with the number α, it is convenient to introduce normalised potentials associated with the radius vector $\boldsymbol{r}_\alpha$ of this particles:

$$\Phi_{lm}^{(0\alpha)}(\boldsymbol{r}) = \frac{\psi_{lm}(\boldsymbol{r}-\boldsymbol{r}_\alpha)}{\|\psi_{lm}(\boldsymbol{r}-\boldsymbol{r}_\alpha)\|}.$$

Scalar product and norm are defined as:

$$(\phi,\psi)_\alpha = \int_{V_\alpha} \nabla\phi^*\nabla\psi \,\mathrm{d}V,$$

$$\|\psi\| = (\psi,\psi)_\alpha^{1/2}, \tag{2}$$

where V_α is the particle volume.

In the case of a cluster of spherical nanoparticles the potential inside each particle is sought as a linear combination of the eigenpotentials of an isolated particle:

$$\Phi^{(\alpha)} = \sum_{ml} A_{lm}^{(\alpha)} \Phi_{lm}^{(0\alpha)}. \tag{3}$$

In (3) and below we use the notation $\sum_{ml} = \sum_{m=-\infty}^{+\infty} \sum_{l=|m|}^{+\infty}$.

The integral equation for the potential inside the particle has the form [1]

$$s\Phi(\boldsymbol{r}) = \frac{1}{4\pi}\int_V \mathrm{d}\boldsymbol{r}'\nabla'\Phi(\boldsymbol{r}')\nabla'\frac{1}{|\boldsymbol{r}-\boldsymbol{r}'|} = \int_V K(\boldsymbol{r},\boldsymbol{r}')\Phi(\boldsymbol{r}')\mathrm{d}V', \tag{4}$$

$$s = \frac{1}{1-\varepsilon/\varepsilon_\mathrm{h}},$$

where integration is performed over the volume V for all the spheres; $\boldsymbol{r},\boldsymbol{r}' \in \bigcup_\alpha V_\alpha$. For the points inside the particle α we obtain from (4):

$$s\Phi^{(\alpha)}(\boldsymbol{r}) = \int_{V_\alpha} K(\boldsymbol{r},\boldsymbol{r}')\Phi^{(\alpha)}(\boldsymbol{r}')\mathrm{d}V'$$

$$+\sum_{\beta\neq\alpha}\int_{V_\beta} K(\boldsymbol{r},\boldsymbol{r}')\Phi^{(\beta)}(\boldsymbol{r}')\mathrm{d}V',\ \boldsymbol{r}\in V_\alpha. \tag{5}$$

For an isolated particle α according to (1) we have

$$\int_{V_\alpha} K(\boldsymbol{r},\boldsymbol{r}')\Phi^{(0\alpha)}(\boldsymbol{r}')\mathrm{d}V' = s_{0l}\Phi^{(0\alpha)},\ \ s_{0l} = \frac{l}{2l+1}. \tag{6}$$

Substituting (3) into (5) and using (6), we obtain

$$(s-s_{0l})\sum_{ml} A_{lm}^{(\alpha)}\Phi_{lm}^{(0\alpha)} = \sum_{\beta\neq\alpha}\int_{V_\beta} K(\boldsymbol{r},\boldsymbol{r}')\sum_{ml} A_{lm}^{(\beta)}\Phi_{lm}^{(0\beta)}\mathrm{d}V'. \tag{7}$$

Multiplying (7) scalarly [see equation (2)] by the function $\Phi_{l'm'}^{(0\alpha)}$ and making a substitution $l'm' \rightleftarrows lm$, we obtain

$$(s-s_{0l})A_{lm}^{(\alpha)} = \sum_{m'l'}\sum_{\beta\neq\alpha} Q_{lml'm'}^{(\alpha,\beta)}A_{l'm'}^{(\beta)},$$

$$l,l' = 0,1,\ldots,\ m=-l\ldots l,\ m'=-l'\ldots l', \tag{8}$$

where

$$Q_{lml'm'}^{(\alpha\beta)} = s_{0l'}(\Phi_{lm}^{(0\alpha)},\Phi_{l'm'}^{(0\beta)})_\alpha$$

$$= s_{0l'}\int_{V_\alpha}\frac{\nabla\psi_{\alpha lm}^{*}\nabla\psi_{\beta l'm'}}{\|\psi_{\alpha lm}\|\|\psi_{\beta l'm'}\|}\mathrm{d}V \tag{9}$$

is the overlap integral of the potentials $\Phi_{lm}^{(0\alpha)}$ and $\Phi_{l'm'}^{(0\beta)}$ of two spheres with the radii a_α and a_β, having the centres at points with the coordinates $\boldsymbol{r}_\alpha$ and $\boldsymbol{r}_\beta$, respectively; $\psi_{\alpha lm}(\boldsymbol{r}) = \psi_{lm}(\boldsymbol{r}-\boldsymbol{r}_\alpha)$; $\psi_{\beta l'm'}(\boldsymbol{r}) = \psi_{l'm'}(\boldsymbol{r}-\boldsymbol{r}_\beta)$. The potential of the particle β outside it can be expanded in series of the eigenfunctions of the particle α:

$$\psi_{\beta l'm'}(\boldsymbol{r}) = \frac{a_\beta^{2l'+1}}{r_{\alpha\beta}^{l+l'+1}}\sum_{l''=0}^{\infty}\sum_{m''=-l''}^{l''}(2l'+1)B_{l''l'm''m'}$$

$$\times\exp[\mathrm{i}(m'-m'')\varphi_\beta]P_{l'+l''}^{m'-m''}(\cos\theta_\beta)\psi_{\alpha l''m''}(\boldsymbol{r}), \tag{10}$$

$$B_{l''l'm''m'} =$$

$$\frac{(-1)^{l'+m'}(l''+l'+m''-m')!}{[(2l'+1)(2l''+1)(l''+m'')!(l''-m'')!(l'+m')!(l'-m')!]^{1/2}},$$

where $r_{\alpha\beta},\theta_\beta,\varphi_\beta$ are the spherical coordinates of the vector $\boldsymbol{r}_{\alpha\beta} = \boldsymbol{r}_\beta - \boldsymbol{r}_\alpha$ in the coordinate system of the particle α. This expression can be obtained, for example, from formulas presented in [38].

Substituting (10) into (9) and using the orthogonality condition of the functions $(\psi_{\alpha lm},\psi_{\alpha l'm'})_\alpha = la_\alpha^{2l+1}\delta_{ll'}\delta_{mm'}$, we find that from the total sum we have only the term with $m''=m$ and $l''=l$:

$$Q_{lml'm'}^{(\alpha\beta)} = \frac{a_\beta^{l'+1/2}a_\alpha^{l+1/2}}{r_{\alpha\beta}^{l+l'+1}}$$

$$\times\exp[\mathrm{i}(m'-m)\varphi_\beta]P_{l'+l}^{m'-m}(\cos\theta_\beta)(ll')^{1/2}B_{ll'mm'}, \tag{11}$$

for other values of the indices the overlap integral is zero.

The system (8), (11) holds for any clusters of spherical particles. In the case of periodically located identical nanoparticles (of radius a) the Bloch theorem is valid, and the expansion coefficients $A_{lm}^{(\alpha)}$ can be expressed as

$$A_{lm}^{(\alpha)} = A_{lm}^{(\boldsymbol{k})}\exp(\mathrm{i}\boldsymbol{k}\boldsymbol{r}_\alpha), \tag{12}$$

where $\boldsymbol{k}$ is the reciprocal lattice vector. Substituting (12) into (8), we obtain

$$(s-s_{0l})A_{lm}^{(\boldsymbol{k})} = \sum_{l'm'} Q_{lml'm'}^{(\boldsymbol{k})}A_{l'm'}^{(\boldsymbol{k})}, \tag{13}$$

where

$$Q_{lml'm'}^{(\boldsymbol{k})} = \sum_{\beta\neq\alpha} Q_{lml'm'}^{(\alpha\beta)}\exp(-\mathrm{i}\boldsymbol{k}\boldsymbol{r}_{\alpha\beta}). \tag{14}$$

In the case of an infinite linear cluster of spherical nanoparticles, expression (14) can be rewritten in the form

$$Q_{lml'm'}^{(\boldsymbol{k})} = \sum_{n\neq n'} Q_{nlmn'l'm'}\exp[-\mathrm{i}(n'-n)kd],$$

$$Q_{nlmn'l'm'} = \left(\frac{a}{r_{\alpha\beta}}\right)^{l+l'+1}\exp[\mathrm{i}(m'-m)\varphi_\beta] \tag{15}$$

$$\times P_{l'+l}^{m'-m}(\cos\theta_\beta)(ll')^{1/2}B_{ll'mm'},$$

where n and n' are the serial numbers counted from the particles located at the origin of the coordinates; $r_{\alpha\beta} = |n'-n|d$ is the distance between the particles, which is determined by their serial numbers (n,n') and the cluster period d. In this case we can assume that $\varphi_\beta = 0$, and $\theta_\beta = 0$ or π depending on the direction of the displacement vector. As a result we obtain

$$Q_{nlmn'l'm'} = \left(\frac{a}{|n'-n|d}\right)^{l+l'+1}$$

$$\times\left(\frac{n'-n}{|n'-n|}\right)^{l+l'}(ll')^{1/2}\delta_{m'm}B_{ll'mm'}. \tag{16}$$

By substituting expression (16) into (15), selecting real and imaginary parts and assuming that the particle α is at the coordinate origin, we find

$$Q^{(\boldsymbol{k})}_{lml'm'} = \begin{cases} \left(\dfrac{a}{d}\right)^{l+l'+1} \displaystyle\sum_{n=1}^{\infty} \frac{\cos(nkd)}{n^{l+l'+1}} K_{ll'm}\delta_{mm'}, \\ \mathrm{i}\left(\dfrac{a}{d}\right)^{l+l'+1} \displaystyle\sum_{n=1}^{\infty} \frac{\sin(nkd)}{n^{l+l'+1}} K_{ll'm}\delta_{mm'} \end{cases} \tag{17}$$

for even and odd $l+l'$, respectively. In (17)

$$K_{ll'm} = 2(-1)^{l'+m'}\left[\frac{ll'}{(2l+1)(2l'+1)}\right]^{1/2} \times \frac{(l+l')!}{\left[(l+m)!(l-m)!(l'+m)!(l'-m)!\right]^{1/2}}.$$

In this case it differs from zero only at $|m| \leqslant l', l$.

One can see from (17) that $Q^{(\boldsymbol{k})}_{lml'm'}$ is diagonal with respect to the azimuthal quantum numbers. Thus, instead of (13) we have a set of systems of equations in which m enters as a parameter:

$$(s - s_{0l})A^{(\boldsymbol{k})}_{lm} = \sum_{l'=|m|}^{\infty} Q^{(\boldsymbol{k})}_{lml'm} A^{(\boldsymbol{k})}_{l'm}, \quad m = 0, 1, \ldots . \tag{18}$$

We will use this system to find the eigenfunctions and eigenfrequencies of the plasmons. Because the obtained solution depends only on the modulus of the vector $\boldsymbol{k}$, we can consider the range $0 \leqslant kd/\pi \leqslant 1$.

Recall that the potential distribution inside the particles, corresponding to each resonance value of the dielectric constant and to each azimuthal number m, is given by

$$\Phi^{(\alpha)}(m) = \sum_{l=|m|}^{\infty} A^{(\alpha)}_{lm} \Phi^{(0\alpha)}_{lm}.$$

The potential outside the cluster is calculated from the integral transform (3) using expressions (4) and (6), modified for external potentials:

$$\Phi(\boldsymbol{r}) = \sum_{\alpha} \Phi^{(\alpha)}_{\mathrm{out}}(\boldsymbol{r}),$$

$$\Phi^{(\alpha)}_{\mathrm{out}}(\boldsymbol{r}) = \sum_{ml} A^{(\alpha)}_{lm} \frac{s_{0l}}{s} \Phi^{(0\alpha)}_{lm}(\boldsymbol{r}) = \sum_{ml} B^{(\alpha)}_{lm} \Phi^{(0\alpha)}_{lm}(\boldsymbol{r}), \ \boldsymbol{r} \in V^{-},$$

where V^{-} is the region outside the cluster and S is the corresponding eigenvalue of (18).

3. Illustrations of the analytical solutions and their comparison with the results of simulations using the finite element method

The infinite system of linear equations (18) was solved numerically taking into account the number of harmonics $l_{\max} = 85$ and the number of neighbours $n_{\max} = 45$. We assumed that the cluster is in a vacuum ($\varepsilon_{\mathrm{h}} = 1$). The increase in $l_{\max}$ and $n_{\max}$ does not lead to any noticeable change in the result. We calculated the resonance values of ε and the eigenvectors corresponding to them. Plasmon frequencies ω of the corresponding modes were determined on the assumption of the classical Drude theory: $\varepsilon(\omega) = 1 - \omega^2_{\mathrm{pl}}/\omega^2$, where ω_{pl} is the the plasma frequency of metal, i.e., $\omega = \omega_{\mathrm{pl}}/\sqrt{1-\varepsilon}$.

Figures 2 and 3 show the resonance frequencies $\omega/\omega_{\mathrm{pl}}$ of plasmons as a function of the distance between the particles

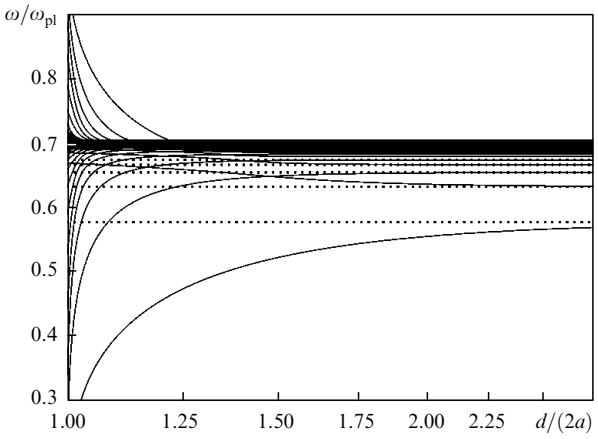

Figure 2. Resonance frequencies $\omega/\omega_{\mathrm{pl}}$ of plasmon oscillations in a linear cluster of spherical nanoparticles as a function of the parameter $d/(2a)$ at $m = 0$ and $k = 0$. Solid curves are the calculations with account for 85 harmonics. Dotted lines correspond to the first five resonance values of $\omega/\omega_{\mathrm{pl}} = \sqrt{l/(2l+1)}$ for a single sphere.

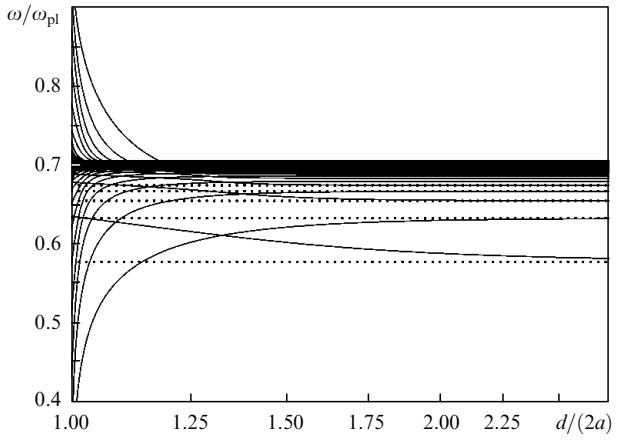

Figure 3. Resonance frequencies $\omega/\omega_{\mathrm{pl}}$ of plasmon oscillations in a linear cluster of spherical nanoparticles as a function of the parameter $d/(2a)$ at $m = 1$ and $k = 0$. Solid curves are the calculations with account for 85 harmonics. Dotted lines correspond to the first five resonance values of $\omega/\omega_{\mathrm{pl}} = \sqrt{l/(2l+1)}$ for a single sphere.

$d/(2a)$. Let us draw attention to the fact that the modes are present both in the low frequency region ($\omega/\omega_{\mathrm{pl}} < 1/\sqrt{2}$), and in the high-frequency region ($1/\sqrt{2} < \omega/\omega_{\mathrm{pl}} < 1$). The latter are called M-type modes and are characterised by strong localisation in the gap between the nanoparticles. They were first discovered relatively recently [21, 39] in studying plasmon resonances in a cluster of two nanoparticles. First we should note that these modes can emerge only at distances that do not exceed $d/(2a) \approx 1.2$, this ratio holding true for an infinite linear cluster and for a cluster of two spheres.

By analogy with the case of two spheres [21, 39] in Figs 2 and 3 we can single out three types of dispersion curves. The modes whose resonance frequency tends to zero with decreasing distance between the particles correspond to antisymmetric plasmon oscillations, i.e., L modes. The modes whose resonance frequency tends to a finite value, which is between 0 and $\omega_{\mathrm{pl}}/\sqrt{2}$ correspond to symmetric plasmon oscillations, i.e., T modes. The modes whose plasmon frequencies tend to ω_{pl} correspond to highly

localised plasmon oscillations, i.e., M modes. Speaking of symmetry, we mean that the symmetry plane is perpendicular to the cluster axis and is in the middle of two nearest spheres.

With increasing the ratio $d/(2a)$ the resonance frequencies of plasmon modes of the cluster undergo transition to the frequencies $\omega/\omega_{\rm pl} = \sqrt{l/(2l+1)}$ ($l = 1, 2, \ldots$ at $m = 0$ and $l \geqslant m$ at $m \neq 0$) of plasmon oscillations of various multipolarity in a single sphere. In the case when $k = 0$ the frequencies of antisymmetric modes tend to the frequencies with odd l at even m and to even l at odd m. The frequencies of symmetric modes transform into the frequency with even l at even m and with odd l at odd m. In the case $kd = \pi$ the situation is similar, but with account for a change in symmetry, i.e., the frequencies of symmetric modes transform into the frequencies with odd l at even m and with even l at odd m, etc.

Consider now the spatial structure of the potential, which corresponds to different modes (Figs 4–9). These potential distributions are obtained by the finite element method in solving the Laplace equation with the help of the Comsol Multiphysics software package. In this case, the resonance values of ε, obtained by solving system (18), are in good agreement with those found in numerical simulations. We also compared the fields inside the particles obtained analytically and calculated using the Comsol software package, and found their good agreement.

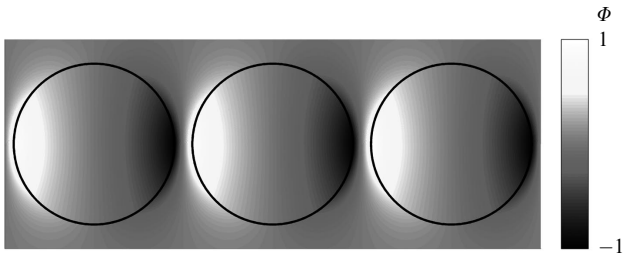

Figure 4. Spatial distribution of the potential Φ_1 of the L_1 mode at $m = 0$, $kd/\pi = 0$, $d/(2a) = 1.11$, $\varepsilon = -5.011$ and $\omega/\omega_{\rm pl} = 0.408$.

In the axially symmetric case ($m = 0$, Figs 4 and 5), the spatial structure of the antisymmetric (L mode) and symmetric (T mode) potentials corresponds on the whole to the structure of the potential of an isolated sphere for $l = 1$ and 2, respectively: $\Phi \sim r^l P_l(\cos\theta)$. Interaction between the spheres in this case is reduced only to a quantitative redistribution of the potential. In this case, the L mode is bright, i.e., has a non-zero (infinite) dipole moment along the cluster axis and can be excited by an external plane wave, while the T mode is dark.

In the case of a symmetric M_1 mode ($m = 0$, Fig. 6) the situation is different – we have a strong concentration of the charge of one sign near the gap between the nanospheres and more or less uniform distribution of the charge of opposite sign in the particle. At points of nanospheres located remotely from the gap, the potential in fact vanishes. Note that for all M modes typical is the localisation of charges near the gap between the spheres. However, unlike the M_1 mode, in the case of M_2 modes both positive and negative charges are concentrated in the gap region.

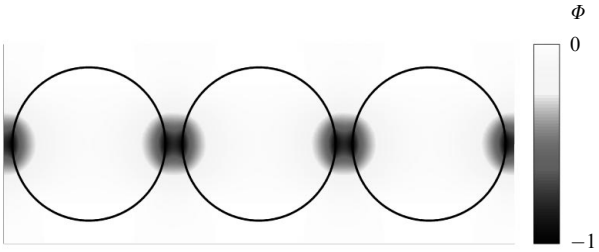

Figure 6. Spatial distribution of the potential Φ of the M_1 mode at $m = 0$, $kd/\pi = 0$, $d/(2a) = 1.11$, $\varepsilon = -0.7721$ and $\omega/\omega_{\rm pl} = 0.751$.

At $m = 1$ (Figs 7 and 8) the situation is similar – the interaction is reduced to the redistribution of the charge. Only in this case the L mode becomes dark and corresponds to the mode with $l = 2$ for one sphere, while the T mode becomes bright with a dipole moment directed along the cluster axis, and corresponds to the mode with $l = 1$ for one sphere. The M modes (Fig. 9) in this case are also bright; however, both positive and negative charges are localised in the region near the gap between the nanospheres.

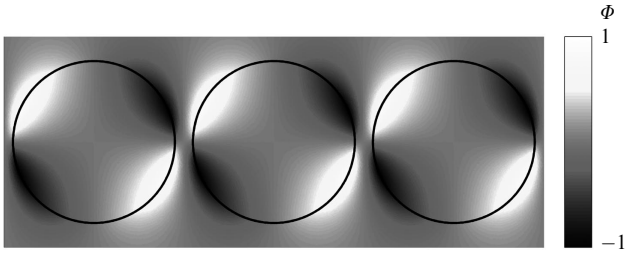

Figure 7. Spatial distribution of the potential Φ of the L_1 mode at $m = 1$, $kd/\pi = 0$, $d/(2a) = 1.11$, $\varepsilon = -2.139$ and $\omega/\omega_{\rm pl} = 0.564$.

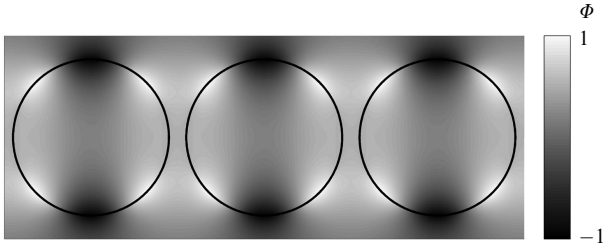

Figure 5. Spatial distribution of the potential Φ_1 of the T_1 mode at $m = 0$, $kd/\pi = 0$, $d/(2a) = 1.11$, $\varepsilon = -1.262$ and $\omega/\omega_{\rm pl} = 0.665$.

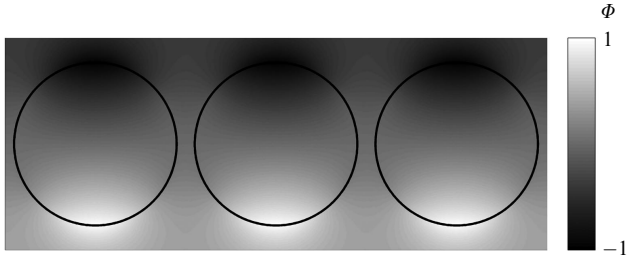

Figure 8. Spatial distribution of the potential Φ of the T_1 mode at $m = 1$, $kd/\pi = 0$, $d/(2a) = 1.11$, $\varepsilon = -1.554$ and $\omega/\omega_{\rm pl} = 0.626$.

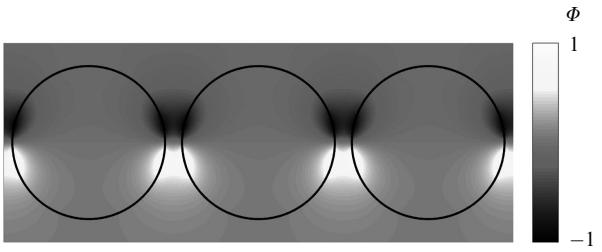

Figure 9. Spatial distribution of the potential Φ of the M_1 mode at $m = 1$, $kd = 0$, $d/(2a) = 1.11$, $\varepsilon = -0.8595$ and $\omega/\omega_{pl} = 0.73$.

With increasing distance between the particles in the cluster the M-mode potential localisation weakens, and in the case of a critical distance between the spheres $d/(2a) \approx 1.2$, M-type localised plasmons disappear, while the L- and T-type plasmons do not experience significant changes, i.e., their spatial structure is preserved and the resonance values of the dielectric constant become equal to known resonance values for a single sphere.

4. Conclusions

We have developed an analytic description of plasmon oscillations of an infinite linear cluster of spherical nanoparticles. We have obtained the dependences of the resonance values of the dielectric constant on the wave number (reciprocal lattice vector) and distance between the spheres in the cluster. We have found their corresponding spatial distribution of the potential. We have observed previously unknown high-frequency modes with strong localisation of the potential in the gap between the cluster particles, i.e., M modes. The results obtained analytically, have been compared with the simulation results by the finite element method using the Comsol Multiphysics software package. We have established their good agreement.

Thus, this work is in fact an exhaustive study of plasmon oscillations of a linear cluster of spherical particles in the quasi-static approximation. Account for the retardation will, of course, affect the results, especially in the frequency region $\omega = ck$, where c is the speed of light. The results of such a study will be given elsewhere.

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