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RESEARCH PAPER

Multi-scattering of electromagnetic waves by nanoshell aggregates

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Abstract A general analytical expression is derived for calculating the total scattering cross-section of an aggregate of nanoparticles. The approach is based on the far zone approximation and repeated use of recursive relations for Bessel functions and Legendre polynomials. In comparison with the existing formula for total scattering cross-section, the expression converges faster and makes it easier to analyze the terms that characterize the interactive coupling between nanoparticles during multiple scattering of electromagnetic waves. The expressions are valid for particle aggregates in which no two particles are in direct contact. Calculated results compare well with measurements for a nanoshell dimer. For a linear chain of particles with the chain axis parallel to the polarization direction of the electric field, analysis shows that the red-shift of resonance peaks results primarily from electrons in metal shells of two adjacent particles oscillating out-of-phase to cancel each other's radiation effects. Multi-scattering in aggregates with more complex arrangements may be explained by combining the effects of linear particle chains.

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Introduction

Nanoshells are a class of nanostructured particles consisting of a nanosized dielectric core and a metal shell, and possess some unique optical properties. One of the most noticeable feature of these nanoshells is that their local surface plasma resonance can be tuned to the frequency of exciting light source (Liu et al. 2008). Use of this property has been explored in various engineering applications including the photothermal therapeutic treatment of cancer patients and/or imaging contrast agents for tumor diagnostics. Many of these applications involve a large number of these nanostructured particles, the optical properties of which are determined by interparticle multiple scattering of light waves (Martin 2006; García de Abajo 1999).

Multiple scattering of electromagnetic waves by a particle aggregate has been widely studied in literature (Liu et al. 2008; Olaofe 1974; Mackowski 1991; Xu 1995; Van de Hulst 1981; Bohren and Huffman 1983; Stratton 1941; Jackson 1976), because of its applications in a broad range of physical, optical, and engineering systems. One critical issue is an accurate prediction of the total scattering cross-section of a particle aggregate. An approach being commonly practiced is to use the scattering cross-section for an isolated particle with its scattering field coefficients replaced by the sum of the scattering field coefficients of each particle in an aggregate (see Eq. (16) below). Research shows that this approach may suffer from two drawbacks for certain configuration of aggregates (Xu 1995). First, a large number of terms often is needed to obtain a reasonably accurate solution for an aggregate of a moderate number of nanoparticles. Second, it is difficult to analyze the behavior of each individual particle and its interaction with others that are responsible for multi-scattering within an aggregate.

This paper presents a new procedure to derive a general analytical expression for the total scattering coefficient of a nanoparticle aggregate, along with validation by available experimental measurements and illustrative examples. This approach overcomes the above two shorting comings and delineates the contribution of interparticle interactions to multi-scattering by a particle aggregate. Numerical experience indicates that the expression presented in this study requires considerably fewer terms to converge and, for certain cases, requires as few as 1/6 terms that are needed in Eq. (16). An analysis of a linear chain of nanoshells illustrates that the red-shift of resonance peaks stems mainly from local coupling of oscillating electrons in surface layers of two adjacent particles. The interactive coupling among nanoshells may be studied also by analyzing the terms of the series in the newly derived expression. An illustrative example of a nanoshell aggregate with a more complicated arrangement is also given.

Mathematical analysis

Consider a multiple nanoparticle system schematically shown in Fig. 1. The incoming wave is assumed to travel in the **k**-direction that forms an angle of α with the z-coordinate and the electric field polarizes linearly in the direction of $\mathbf{e}_E = -\cos\alpha \cos\gamma\ \mathbf{e}_x + \sin\gamma\ \mathbf{e}_y + \sin\alpha \cos\gamma\ \mathbf{e}_z$. It is known that the electromagnetic waves are scattered by the nanoparticle aggregate in a complex fashion. An income wave is scattered by one particle in the aggregate and its scattered wave then becomes an income wave to other particles and so on. These complex multi-scattering phenomena are described by the Maxwell equations, which for the problem under consideration, reduce to the vector wave equations,

$$\nabla^2\mathbf{E} + k^2\mathbf{E} = 0, \quad \nabla \cdot \mathbf{E} = 0, \\ \mathbf{H} = -(i/\mu_0\omega)\nabla \times \mathbf{E} \tag{1}$$

where **E** is the electric field, **H** the magnetic intensity, $k = (\varepsilon\mu_0\omega)^{1/2}$ the wave number, ε_0 the permittivity, μ_0 the magnetic permeability, and ω the frequency of the applied field.

For a single, isolated spherical particle with excitation by a plane wave as shown in Fig. 1, the solution for the scattered electromagnetic field may be obtained by the multipole expansion technique (Stratton 1941; Jackson 1976),

$$\begin{aligned} \mathbf{E}_s &= \sum_{n=1}^{\infty}\sum_{m=-n}^{m=n}(a_{mn}\mathbf{m}_{mn} + b_{mn}\mathbf{n}_{mn}); \\ \mathbf{H}_s &= -(i/\omega\mu)\nabla \times \mathbf{E}_s \\ &= -\frac{ik}{\omega\mu}\sum_{n=1}^{\infty}\sum_{m=-n}^{m=n}(b_{mn}\mathbf{m}_{mn} + a_{mn}\mathbf{n}_{mn}) \end{aligned} \tag{2}$$

where the vector wave functions are defined as follows,

$$\begin{aligned} \mathbf{m}_{mn} &= i\pi_{mn}(\cos\theta)z_n(kr)e^{im\varphi}\mathbf{e}_\theta \\ &\quad - z_n(kr)\tau_{mn}(\cos\theta)e^{im\varphi}\mathbf{e}_\varphi \end{aligned} \tag{3}$$

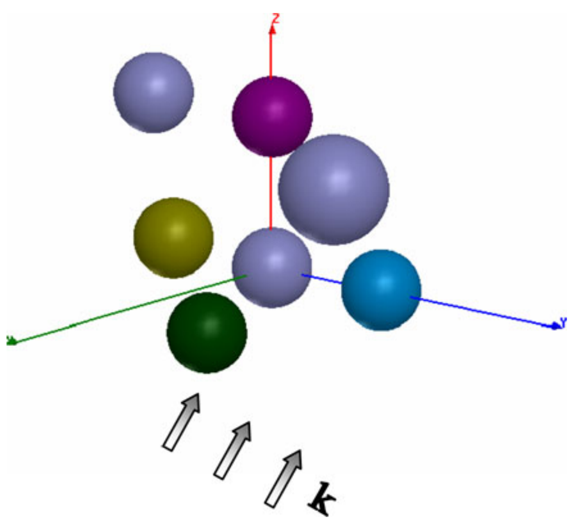

Fig. 1 Schematic of a multiple particle system

$$\mathbf{n}_{mn} = \frac{n(n+1)}{kr} z_n(kr) P_n^m(\cos\theta) e^{im\varphi} \mathbf{e}_r + \frac{1}{kr} \frac{\partial[r z_n(kr)]}{\partial r} \tau_{mn}(\cos\theta) e^{im\varphi} \mathbf{e}_\theta + \frac{i}{kr} \frac{\partial[r z_n(kr)]}{\partial r} \pi_{mn}(\cos\theta) e^{im\varphi} \mathbf{e}_\varphi \tag{4}$$

with $z_n(kr) = h^{(1)}(kr)$ for the scattering field and two auxiliary functions,

$$\pi_{mn}(\cos\theta) = \frac{m}{\sin\theta} P_n^m(\cos\theta); \quad \tau_{mn}(\cos\theta) = \frac{d}{d\theta} P_n^m(\cos\theta) \tag{5}$$

In the above equation, $P_n^m(\cos\theta)$ is the associate Legendre polynomial,

$$P_t^s(\mu) = (1-\mu^2)^{s/2} \frac{dP_t(\mu)}{d\mu} \tag{6}$$

The coefficients in the scattering field, a_{mn} and b_{mn}, are calculated by the following expressions,

$$a_{mn}^i = p_{mn}^i \frac{\psi(m^i x^i)\psi'(x^i) - m^i \psi'(m^i x^i)\psi(x^i)}{\psi(m^i x^i)\xi'(x^i) - m^i \psi'(m^i x^i)\xi(x^i)} = -p_{mn}^i \alpha^i;$$
$$b_{mn}^i = q_{mn}^i \frac{m^i \psi(m^i x^i)\psi'(x^i) - \psi'(m^i x^i)\psi(x^i)}{m^i \psi(m^i x^i)\xi'(x^i) - \psi'(m^i x^i)\xi(x^i)} = -q_{mn}^i \beta^i \tag{7}$$

where p_{mn} and q_{mn} are calculated by the following expression,

$$p_{mn} = -E_0 \frac{2n+1}{n(n+1)} \frac{(n-m)!}{(n+m)!} i^n \times \left(\pi_{mn}(\alpha)\sin\gamma - i\tau_{mn}(\alpha)\cos\gamma \right);$$
$$q_{mn} = E_0 \frac{2n+1}{n(n+1)} \frac{(n-m)!}{(n+m)!} i^n \times \left(i\pi_{mn}(\alpha)\cos\gamma - \tau_{mn}(\alpha)\sin\gamma \right)$$

where α is the angle between the z-axis and the wave vector $\mathbf{k}$ (Xu 1995) and E_0 is the magnitude of the electric field. Also, $m^i = k^i/k$ and $k^i = (\varepsilon^i \mu_0 \omega)^{1/2}$, with the superscript i denoting the ith sphere in the nanoparticle aggregate, and k for the medium surrounding the particle; and $m^i x^i = k^i a^i$, a^i being the radius of the ith particle.

A simpler form of p_{mn} and q_{mn} is obtained if the electric field travels in the z-direction and polarizes in the x-direction, viz.,

$$p_{1,n}^i = -0.5 E_0 i^{n+1} \frac{2n+1}{n(n+1)} \exp[ikZ^i] = q_{1,n}^i; \quad p_{-1,n} = 0.5 E_0 i^{n+1} (2n+1) \exp[ikZ^i] = -q_{-1,n} \tag{8}$$

where Z is the z-position of the center of a sphere in global coordinates.

For multi-scattering of electromagnetic waves by an aggregate of nanoparticles, a general framework is developed via the vector addition theorem (Stein 1961; Cruzan 1962). A crucial step of the theorem involves translating the coordinate systems for each sphere to a common coordinate system. To preserve the spherical coordinate system, it is often convenient to choose a reference sphere whose center is the origin of the common coordinate system. Then, the spherical coordinate system attached to each sphere is translated to the reference system. The translation rule for the vector wave functions describing the electric and magnetic fields is discussed in two widely cited references in scattering literature and takes the following forms (Stein 1961; Cruzan 1962),

$$\mathbf{m}_{mn} = \sum_{\mu\nu} \left(A_{\mu\nu}^{mn} \mathbf{m}_{\mu\nu} + B_{\mu\nu}^{mn} \mathbf{n}_{\mu\nu} \right); \quad \mathbf{n}_{mn} = \sum_{\mu\nu} \left(B_{\mu\nu}^{mn} \mathbf{m}_{\mu\nu} + A_{\mu\nu}^{mn} \mathbf{n}_{\mu\nu} \right) \tag{9}$$

$$A_{\mu\nu}^{mn} = (-1)^\mu i^{\nu-n} \frac{2\nu+1}{2\nu(\nu+1)} \sum_{p=|n-\nu|}^{p=n+\nu} i^p a(m,n;-\mu,\nu;p)[n(n+1)+\nu(\nu+1) - p(p+1)] z_p(kr_0) P_p^{m-\mu}(\cos\theta_0) e^{i(m-\mu)\varphi_0} \tag{10}$$

$$B_{\mu\nu}^{mn} = (-1)^\mu i^{\nu-n} \frac{(2\nu+1)}{2\nu(\nu+1)} \sum_{p=|n-\nu|+1}^{p=n+\nu+1} i^p [(n+\nu+1)^2 - p^2][p^2-(n-\nu)^2] b(m,n;-\mu,\nu;p,p-1) z_p(kr_0) P_p^{m-\mu}(\cos\theta_0) e^{i(m-\mu)\varphi_0} \tag{11}$$

where $z_p(kr_0) = h_p(kr_0)$ and

$$a(m,n,\mu,v,p) = (-1)^{m+\mu}\frac{2p+1}{2}\frac{(p+m+\mu)!}{(p-m-\mu)!}$$
$$\int_{-1}^{1} P_n^m P_v^\mu P_p^{m+\mu} dx \ = (-1)^{m+\mu}(2p+1)$$
$$\sqrt{\frac{(n+m)!}{(n-m)!}\frac{(v+\mu)!}{(v-\mu)!}\frac{(p-m-\mu)!}{(p+m+\mu)!}}$$
$$\begin{pmatrix} n & v & p \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} n & v & p \\ m & \mu & -m-\mu \end{pmatrix} \tag{12}$$

$$a(m,n,\mu,v,p,q) = (-1)^{-m-\mu}(2p+1)$$
$$\sqrt{\frac{(n+m)!}{(n-m)!}\frac{(v+\mu)!}{(v-\mu)!}\frac{(p-m-\mu)!}{(p+m+\mu)!}}$$
$$\begin{pmatrix} n & v & q \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} n & v & p \\ m & \mu & -m-\mu \end{pmatrix} \tag{13}$$

where $\begin{pmatrix} \times & \times & \times \\ \times & \times & \times \end{pmatrix}$ is the Wigner 3-j symbols.

Having determined the translation rule for the vector field, the coefficients in new coordinates are calculated by matching the boundary conditions of the electric and magnetic fields at the surface of the reference sphere. The procedure yields the expressions for the coefficients,

$$\begin{aligned} a^i_{\pm 1,n} &= -\alpha^i(q^i_{\pm 1,n} + Q_{\pm 1.n}(i)); \\ b^i_{\pm 1,n} &= -\beta^i(p^i_{\pm 1,n} + R_{\pm 1,n}(i)) \end{aligned} \tag{14}$$

$$Q_{mn}(i) = \sum_{\substack{j=1 \\ j\neq i}}^{N}\sum_{l=1}^{\infty}\sum_{k=-l}^{l}\left(a^j_{kl}A^{kl}_{mn}(ji) + b^j_{kl}B^{kl}_{mn}(ji)\right);$$
$$R_{mn}(i) = \sum_{\substack{j=1 \\ j\neq i}}^{N}\sum_{l=1}^{\infty}\sum_{k=-l}^{l}\left(b^j_{kl}A^{kl}_{mn}(ji) + a^j_{kl}B^{kl}_{mn}(ji)\right) \tag{15}$$

where the subscript i, j refer to the ith and jth spheres, respectively, and (ji) refers to the coordinate transformation from the jth sphere to the ith sphere.

It is noted that the above equations result in a system of equations with the unknowns being the scattering coefficients for each of the sphere in the aggregate, and these coefficients are linked by the transformation matrix. The detailed structure and solution procedures may be found in (Mackowski 1991) and thus are omitted.

A common procedure for calculating the scattering coefficient of a nanoparticle aggregate basically involves two steps: (1) choose a reference sphere and translate the coordinate system of each sphere to that of the reference sphere and (2) integrate the Poynting vector of the scattered electromagnetic fields contributed by all the spheres over a large spherical surface far away from the center of the reference coordinates defined by the reference sphere so that the surface encloses all the nanoparticles. This procedure then yields the following expression for the scattering coefficients,

$$C_{sca} = \frac{4\pi}{k^2}\sum_{l}^{L}\sum_{n}^{\infty}\sum_{m=-n}^{m=n}\frac{n(n+1)}{2n+1}\frac{(n+m)!}{(n-m)!}\left(a^T_{mn}a^{*T}_{mn} + b^T_{mn}b^{*T}_{mn}\right) \tag{16}$$

where

$$a^T_{mn} = a^1_{mn} + R_{mn}(1);\ b^T_{mn} = b^1_{mn} + Q_{mn}(1)$$
$$Q_{mn}(1) = \sum_{\substack{j=1 \\ j\neq i}}^{N}\sum_{l=1}^{\infty}\sum_{k=-l}^{l}\left(a^j_{kl}A^{kl}_{mn}(j1) + d^j_{kl}B^{kl}_{mn}(j1)\right);$$
$$R_{mn}(1) = \sum_{\substack{j=1 \\ j\neq i}}^{N}\sum_{l=1}^{\infty}\sum_{k=-l}^{l}\left(b^j_{kl}A^{kl}_{mn}(j1) + a^j_{kl}B^{kl}_{mn}(j1)\right)$$

with $z_p(kr) = j_p(kr)$ used in A^{kl}_{mn} and B^{kl}_{mn}.

It is now commonly accepted that there are at least two problems with this complex expression (Xu 1995). First of all, there is no specific knowledge of how many terms are needed to obtain a solution with a required accuracy. Numerical tests conducted in our laboratories and others' indicate that the terms required for an accurate solution depends upon the separation distance between the particles. For a two particle system, for instance, the terms required for an accurate solution increase rapidly as the particles become further apart. When the particles are separated far away so that their interactions are negligible, the total scattering cross-section will be the sum of the cross-sections of each of the individual particles. To recover that, however, an extremely large number of terms needs to be used, which can be computationally intensive, even with current powerful computing platform. Another problem is that the interactive nature of multiple particles in an aggregate is sealed in the complex expression and thus makes it difficult to identify the contributions from each of the particles in the aggregate. From the purpose of theoretical analysis, it becomes rather difficult with using Eq. (16) to identify the terms governing the interparticle-scattering in an aggregate of nanoparticles.

As the surface integration is carried out outside the particle aggregate, it is thus possible to extend this surface to the far zone and use the asymptotic behavior of the Bessel and Hankel functions to facilitate the integration of the radiation fields. Toward this end, we consider the far zone approximation of the Hankel function (Jackson 1976),

$$h_n(kr) = (-i)^{n+1}\frac{e^{ikr}}{kr} \tag{17}$$

With the above substituted into Eq. (9), the following expressions are obtained for the **m** and **n** vector functions in far zone,

$$\begin{aligned}\mathbf{m}_{mn}^{(3)} &= (-i)^n[\mathbf{e}_\theta\pi_{mn}(\cos\theta) \\ &\quad + i\mathbf{e}_\varphi\tau_{mn}(\cos\theta)]\frac{e^{ikr}}{kr}e^{im\varphi}; \\ \mathbf{n}_{mn}^{(3)} &= (-i)^n[\mathbf{e}_\theta\tau_{mn}(\cos\theta) + i\mathbf{e}_\varphi\pi_{mn}(\cos\theta)]\frac{e^{ikr}}{kr}e^{im\varphi}\end{aligned} \tag{18}$$

Consequently, the scattering electric and magnetic fields take the forms of,

$$\begin{aligned}\mathbf{E} &= \sum_n^\infty\sum_{m=-n}^{m=n}(a_{mn}^T\mathbf{N}_{mn}^{(3)} + b_{mn}^T\mathbf{M}_{mn}^{(3)}) \\ &= \frac{e^{ikr}}{r}\sum_n^\infty(-i)^n\sum_{m=-n}^{m=n}e^{im\varphi}\{\mathbf{e}_\theta[a_{mn}^T\tau_{mn} + b_{mn}^T\pi_{mn}] \\ &\quad + i\mathbf{e}_\varphi[a_{mn}^T\pi_{mn} + b_{mn}^T\tau_{mn}]\}\end{aligned} \tag{19}$$

$$\begin{aligned}\mathbf{H} &= -\frac{ik}{\omega\mu}\sum_n^\infty\sum_{m=-n}^{m=n}(b_{mn}^T\mathbf{N}_{mn}^{(3)} + a_{mn}^T\mathbf{M}_{mn}^{(3)}) \\ &= -\frac{ik}{\omega\mu}\frac{e^{ikr}}{kr}\sum_n^\infty(-i)^n\sum_{m=-n}^{m=n}e^{im\varphi}\{\mathbf{e}_\theta[b_{mn}^T\tau_{mn} \\ &\quad + a_{mn}^T\pi_{mn}] + \mathbf{e}_\varphi[b_{mn}^T\pi_{mn} + a_{mn}^T\tau_{mn}]\}\end{aligned} \tag{20}$$

In the far zone approximation, however, the coefficients may also take a simpler form (Xu 1995),

$$a_{mn}^T = \sum_l^L e^{-ik\mathbf{d}_l\cdot\mathbf{e}_r}a_{mn}^l; \quad b_{mn}^T = \sum_l^L e^{-ik\mathbf{d}_l\cdot\mathbf{e}_r}b_{mn}^l \tag{21}$$

where $\boldsymbol{d}_l = \mathbf{r} - \mathbf{r}_l$ is the vector pointing from the center of the lth sphere to the point of observation **r**. The total scattering cross-section is calculated by integrating the Poynting vector over a spherical surface enclosing the ensemble of the particles, viz.,

$$C_{\text{sca}} = \frac{0.5}{I_0}\int_{4\pi} r^2\mathbf{E}\times\mathbf{H}^*\cdot\mathbf{e}_r d\Omega \tag{22}$$

where $I_0 = k|E_0|^2/(2\omega\mu_0)$, the superscript * denotes the complex conjugate and r is the radius of the spherical surface for integration. The time-averaged Poynting vector may be expanded explicitly. With the far zone approximation for the coefficients a^T and b^T substituted, the following expression for the integrand is obtained,

$$\begin{aligned}r^2\mathbf{E}\times\mathbf{H}^*\cdot\mathbf{e}_r &= \left(\frac{e^{ikr}}{kr}\sum_n^\infty(-i)^n\sum_{m=-n}^{m=n}e^{im\varphi}\{\mathbf{e}_\theta[a_{mn}^T\tau_{mn} + b_{mn}^T\pi_{mn}] + i\mathbf{e}_\varphi[a_{mn}^T\pi_{mn} + b_{mn}^T\tau_{mn}]\}\right) \\ &\quad\times\left(\frac{ik}{\omega\mu_0}\frac{e^{-ikr}}{kr}\sum_v^\infty(i)^v\sum_{\mu=-v}^{\mu=v}e^{-i\mu\varphi}\left\{\mathbf{e}_\theta\left[b_{\mu v}^{*T}\tau_{\mu v} + a_{\mu v}^{*T}\pi_{\mu v}\right] - i\mathbf{e}_\varphi\left[a_{mn}^{*T}\tau_{mn} + b_{mn}^{*T}\pi_{mn}\right]\right\}\right) \\ &= \frac{1}{\omega\mu_0 k}\sum_n^\infty\sum_v^\infty(-1)^n i^{n+v}\sum_{m=-n}^{m=n}\sum_{\mu=-v}^{\mu=v}e^{i(m-\mu)\varphi}\sum_l^L[(a_{mn}^l a_{\mu v}^{*l} + b_{mn}^l b_{\mu v}^{*l})(\tau_{mn}\tau_{\mu v} + \pi_{mn}\pi_{\mu v}) \\ &\quad + (b_{mn}^l a_{\mu v}^{*l} + a_{mn}^l b_{\mu v}^{*l})(\pi_{mn}\tau_{\mu v} + \tau_{mn}\pi_{\mu v})] \\ &\quad + \frac{1}{\omega\mu_0 k}\sum_n^\infty\sum_v^\infty(-1)^n i^{n+v}\sum_{m=-n}^{m=n}\sum_{\mu=-v}^{\mu=v}e^{i(m-\mu)\varphi}\sum_l^L\sum_{\lambda\neq l}^L e^{ik\mathbf{u}_{\lambda l}\cdot\mathbf{e}_r}[(a_{mn}^l a_{\mu v}^{*\lambda} + b_{mn}^l b_{\mu v}^{*\lambda})(\tau_{mn}\tau_{\mu v} + \pi_{mn}\pi_{\mu v}) \\ &\quad + (b_{mn}^l a_{\mu v}^{*\lambda} + a_{mn}^l b_{\mu v}^{*\lambda})(\pi_{mn}\tau_{\mu v} + \tau_{mn}\pi_{\mu v})]\end{aligned} \tag{23}$$

where $\boldsymbol{u}_{\lambda l} = \mathbf{d}_\lambda - \mathbf{d}_l$, which represents the displacement between two particles λ and l.

Integration of the first two terms is relatively straightforward by the use of the orthoganality relations for the associate Legendre polynomials (Stratton 1941),

$$\int_{4\pi} e^{i(m-\mu)\varphi}(\tau_{mn}\tau_{\mu v} + \pi_{mn}\pi_{\mu v})d\Omega = \frac{4\pi n(n+1)}{2n+1}\frac{(n+m)!}{(n-m)!}\delta_{nv}\delta_{m\mu} \tag{24}$$

Integration of the last two terms containing the factor$e^{ik\mathbf{u}_{\lambda l}\cdot\mathbf{e}_r}$is more involved. To facilitate the calculation, the following spherical wave function expansion is considered (Jackson 1976),

$$e^{ik\mathbf{u}_{\lambda l}\cdot\mathbf{e}_r} = \sum_t^{\infty}\sum_{s=-t}^{t}(-1)^s(2t+1)i^t j_t(ku_{\lambda l})P_t^{-s}(\cos\theta_{\lambda l})e^{-is\varphi_{\lambda l}}P_t^s(\cos\theta)e^{is\varphi} \tag{25}$$

With this, the following type of integral appears,

$$\int_{4\pi} e^{i(s+m-\mu)\varphi}(\tau_{mn}\tau_{\mu v} + \pi_{mn}\pi_{\mu v})P_t^s d\Omega = 2\pi\int_{-1}^{1}\left(\frac{m\mu P_n^m P_v^\mu}{(1-x^2)} + (1-x^2)\frac{dP_n^m}{dx}\frac{dP_v^\mu}{dx}\right)P_t^s dx \tag{26}$$

The above integral is zero if $s + m - \mu \neq 0$. To integrate the product of the associate Legendre polynomials, we consider the general Legendre differential equation for P_n^m,

$$\frac{d}{dx}\left((1-x^2)\frac{dP_n^m}{dx}\right) + \left(n(n+1) - \frac{m^2}{1-x^2}\right)P_n^m = 0 \tag{27}$$

Multiplying by $P_v^\mu P_t^s$ and then integrating by parts, one has the following relation for P_n^m with $P_v^\mu P_t^s$,

$$\int_{-1}^{1}(1-x^2)\frac{dP_n^m}{dx}\frac{dP_v^\mu P_t^s}{dx}dx = n(n+1)\int_{-1}^{1}P_n^m P_v^\mu P_t^s dx - m^2\int_{-1}^{1}\frac{P_n^m P_v^\mu P_t^s}{1-x^2}dx \tag{28}$$

The same procedure can be applied to obtain the two similar relations for P_v^μ with $P_n^m P_t^s$ and for P_t^s with $P_n^m P_v^\mu$, respectively. Adding the relations for P_n^m with $P_v^\mu P_t^s$and for P_v^μ with $P_n^m P_t^s$, then subtracting from the result the relation for P_t^s with $P_n^m P_v^\mu$, and re-arranging, one has the following result,

$$2m\mu\int_{-1}^{1}\frac{P_n^m P_v^\mu P_t^s}{1-x^2}dx + 2\int_{-1}^{1}(1-x^2)P_t^s\frac{dP_v^\mu}{dx}\frac{dP_n^m}{dxdx} = [n(n+1) + v(v+1) - t(t+1)]\int_{-1}^{1}P_n^m P_v^\mu P_t^s dx \tag{29}$$

where the condition $s + m - \mu = 0$ has been used.

With the relations for the associate Legendre polynomials,

$$P_n^m P_v^\mu = \sum_{p=|n-v|}^{n+v} a(m,n,\mu,v,p)P_p^{\mu+m};\quad P_n^m = (-1)^m\frac{(n+m)!}{(n-m)!}P_n^{-m};\quad \int_{-1}^{1}P_p^s P_t^s dx = \frac{2}{2p+1}\frac{(p+s)!}{(p-s)!}\delta_{pt} \tag{30}$$

where $a(m, n, \mu, v, p)$ is calculated using Eq. (12), the triple product can be integrated with the result,

$$\int_{-1}^{1}P_n^m P_v^\mu P_t^s dx = (-1)^{-m}\frac{(n+m)!}{(n-m)!}\int_{-1}^{1}P_n^{-m}P_v^\mu P_t^s dx = (-1)^{-m}\frac{(n+m)!}{(n-m)!}\sum_{p=|n-v|}^{n+v}a(-m,n,\mu,v,p)\frac{2}{2p+1}\frac{(p+s)!}{(p-s)!}\delta_{pt} \tag{31}$$

Here, $a(-m, n, \mu, v, p)$ is given by Eq. (13).

Integration of the last term in Eq. (23) yields the result,

$$\int_{4\pi} e^{i(s+m-\mu)\varphi}(\tau_{mn}\pi_{\mu v} + \pi_{mn}\tau_{\mu v})P_t^s d\Omega = -2\pi\int_{-1}^{1}\left(mP_n^m\frac{dP_v^\mu}{dx} + \mu P_v^\mu\frac{dP_n^m}{dx}\right)P_t^s dx \tag{32}$$

With the recursive relations for the associate Legendre polynomials,

$$\frac{dP_n^m}{dx} = \frac{-mxP_n^m}{1-x^2} + \frac{P_n^{m+1}}{\sqrt{1-x^2}};$$
$$\frac{dP_v^\mu}{dx} = \frac{\mu x P_v^\mu}{1-x^2} - (v-\mu+1)(v+\mu)\frac{P_v^{\mu-1}}{\sqrt{1-x^2}} \tag{33}$$

the integrand becomes

$$\begin{aligned}
&\mu P_t^s P_v^\mu \frac{dP_n^m}{dx} + mP_t^s P_n^m \frac{dP_v^\mu}{dx} = \frac{mP_t^s P_v^{\mu+1} P_n^m}{\sqrt{1-x^2}} \\
&- \mu(n-m+1)(n+m)\frac{P_t^s P_n^{m-1} P_v^\mu}{\sqrt{1-x^2}} \\
&= (-1)^m \frac{(n+m)!}{(n-m)!} \sum_{p=|n-v|}^{n+v} [ma(-m,n,\mu+1,v,p) \\
&+ \mu a(-m+1,n,\mu,v,p)] \frac{P_t^s P_p^{s+1}}{\sqrt{1-x^2}}
\end{aligned} \tag{34}$$

where $s = \mu - m$ and use has been made of the following relations,

$$P_v^\mu P_{n+1}^{-m} = \sum_{p=|n-v+1|}^{p=v+n+1} a(\mu, v, -m, n+1, p) P_p^s \tag{35}$$

$$P_n^m = (-1)^{-m} \frac{(n+m)!}{(n-m)!} P_n^{-m} \tag{36}$$

Making repeated use of another recursive relation for the associated Legendre polynomials,

$$\frac{P_{n+1}^{m+1}}{\sqrt{1-x^2}} = (2n+1)P_n^m + \frac{P_{n-1}^{m+1}}{\sqrt{1-x^2}} \tag{37}$$

yields the following relation,

$$\frac{P_t^s P_p^{s+1}}{\sqrt{1-x^2}} = \sum_{q=1,3,}^{q=n-s} (2n-2q+1) P_t^s P_p^s; \quad s \geq 0 \tag{38}$$

Eq. (38) now can be integrated with the result,

$$\int_{-1}^{1} \frac{P_t^s P_p^{s+1}}{\sqrt{1-x^2}} dx = (1 - (-1)^{t+p}) \sum_{q=1,3,}^{q=p-s} \frac{(t+s)!}{(t-s)!} \delta_{t,p-q}; \quad s \geq 0 \tag{39}$$

For the case of $s < 0$, Eq. (36) is applied first, followed by the use of Eqs. (37) and (38). Integration then yields the result,

$$\int_{-1}^{1} \frac{P_t^s P_p^{s+1}}{\sqrt{1-x^2}} dx = (1 - (-1)^{t+p}) \sum_{q=1,3,}^{q=t-s} \frac{(t+s)!}{(t-s)!} \delta_{t,p-q}; \ s < 0 \tag{40}$$

With Eqs. (25–40) and after lengthy but elementary algebraic manipulation, the analytical expression for the scattering cross-section for a multiparticle system is obtained below,

$$\begin{aligned}
&C_{sca} \\
&= \frac{4\pi}{k^2} \sum_n^\infty \sum_{m=-n}^{n} \sum_l^L \Bigg((a_{mn}^l a_{mn}^{*l} + b_{mn}^l b_{mn}^{*l}) \\
&\quad \times \frac{n(n+1)}{2n+1} \frac{(n+m)!}{(n-m)!} \\
&\quad + \mathrm{Re} \sum_v^\infty \sum_{\mu=-v}^{\mu=v} \sum_{\lambda \neq l}^L \Big[\Big(a_{mn}^l a_{\mu v}^{*\lambda} + b_{mn}^l b_{\mu v}^{*\lambda} \Big) C_{mn\mu v} \\
&\quad + (b_{mn}^l a_{\mu v}^{*\lambda} + a_{mn}^l b_{\mu v}^{*\lambda}) D_{mn\mu v} \Big] \Bigg)
\end{aligned} \tag{41}$$

where $C_{mn\lambda v}$ is obtained by combining Eqs. (25), (26), and (31),

$$\begin{aligned}
C_{mn\mu v} &= \frac{(-1)^n i^{n+v}}{4\pi} \sum_{t=0}^{\infty} \sum_{s=-t}^{t} (-1)^s (2t+1) i^t j_t(ku_{\lambda l}) \\
&\quad \times P_t^{-s}(\cos\theta_{\lambda l}) e^{-is\varphi_{\lambda l}} \int_0^{2\pi} e^{i(s+m-\mu)} d\varphi \\
&\quad \times \int_{-1}^{1} (\tau_{mn}\tau_{\mu v} + \pi_{mn}\pi_{\mu v}) P_t^s dx \\
&= \frac{(-1)^{\mu+n}}{2} \frac{(n+m)!}{(n-m)!} \\
&\quad \times \sum_{p=|n-v|}^{n+v} i^{p+n+v} [n(n+1) + v(v+1) \\
&\quad - p(p+1)] \frac{(p-m+\mu)!}{(p+m-\mu)!} a(-m,n,\mu,v,p) \\
&\quad \times j_p(ku_{\lambda l}) P_p^{m-\mu}(\cos\theta_{\lambda l}) e^{i(m-\mu)\varphi_{\lambda l}}
\end{aligned} \tag{42}$$

and $D_{mn\lambda v}$ by Eqs. (25), (34), (41), and (42),

$$D_{mn\mu v} = \frac{(-1)^n i^{n+v}}{4\pi} \sum_{t=0}^{\infty} \sum_{s=-t}^{t} (-1)^s (2t+1) i^t j_t(ku_{\lambda l})$$
$$\times P_t^{-s}(\cos\theta_{\lambda l}) e^{-is\varphi_{\lambda l}} \int_0^{2\pi} e^{i(s+m-\mu)} d\varphi$$
$$\times \int_{-1}^{1} (\pi_{mn}\tau_{\mu v} + \tau_{mn}\pi_{\mu v}) P_t^s dx$$
$$= \frac{(-1)^{n+\mu}}{2} \frac{(n+m)!}{(n-m)!}$$
$$\times \sum_{p=|n-v|}^{n+v} \sum_{t=|\mu-m|}^{\infty} (2t+1) i^{t+n+v} d(m,n,\mu,v,p,t)$$
$$\times j_t(ku_{\lambda l}) P_t^{m-\mu}(\cos\theta_{\lambda l}) e^{i(m-\mu)\varphi_{\lambda l}} \tag{43}$$

with

$$d(m,n,\mu,v,p,t)$$
$$= ((-1)^{t+p} - 1)\operatorname{sgn}(\mu - m + \tfrac{1}{2})$$
$$\times H((p-t)(\mu - m + \tfrac{1}{2})) \tag{44}$$
$$\times [ma(-m,n,\mu+1,v,p)$$
$$+ \mu a(-m+1,n,\mu,v,p)] \frac{(t+\mu-m)!}{(t-\mu+m)!}$$

and

$$H(x) = \begin{cases} 0; & x<0 \\ 1; & x \geq 0 \end{cases}$$

The scattering cross-section having been determined, one can compute the total scattering coefficient of the aggregate defined by the cross-section of the reference sphere,

$$Q_{sca} = C_{sca}/\pi a^2 \tag{45}$$

Results and discussion

The physical meaning of Eq. (41) for multiple scattering is now clear. The first term represents the independent scattering of electromagnetic waves by individual particles, whereas the second stands for the interaction between any two different particles in the multiparticle system. This physical picture is not clear directly with Eq. (16). If the particles are isolated, that is, the particles do not interfere with each other during scattering, the second series makes no contribution to scattering. This happens when the particles are separated in a sufficiently large distance. In theory, when the particles are apart at infinity, their scattering behavior is independent of each other. To see that, we may expand the Bessel function in its asymptotic form,

$$j_t(ku_{\lambda l}) \sim \frac{2}{\pi k u_{\lambda l}} \cos(ku_{\lambda l} - 0.5t\pi - 0.25\pi) \text{ as } u_{\lambda l} \to \infty \tag{46}$$

Since now all the terms are bounded, the second series in Eq. (41) vanishes as $u_{\lambda l} \to \infty$ and the case of independent scattering is recovered.

Perhaps, the simplest system of all is a two particle configuration. For this case, one can always select the z-axis along the line connecting the centers of the two spheres. Thus, a simplified expression is obtained by substituting the parameters $d = u_{\lambda l}\,\theta_{\lambda l} = 0$, $s = 0$ into Eq. (41). Alternatively, it can be shown that for a linear chain of particles,

$$e^{ik\mathbf{u}_{\lambda l}\cdot\mathbf{e}_r} = e^{\pm ikd_{\lambda l}\cos\theta}$$
$$= \sum_{t=0}^{\infty} (\pm i)^t (2t+1) j_t(kr) P_t(\cos\theta) \tag{47}$$

The above procedure used in Eqs. (26–41) yields the following simplified expression for the total scattering cross-section of the linear chain,

$$C_{sca}$$
$$= \frac{2\pi k}{2\omega\mu_0 I_0} \sum_n^{\infty} \sum_{m=-n}^{n} \sum_l^{L} \Big((a_{mn}^l a_{mn}^{*l} + b_{mn}^l b_{mn}^{*l})$$
$$\times \frac{2}{2n+1} \frac{(n+m)!}{(n-m)!}$$
$$+ \operatorname{Re} \sum_v^{\infty} (-1)^{n+v} i^{n+v} \sum_{\lambda \neq l}^{L} (C_{mnv}(a_{mn}^l a_{mv}^{*\lambda} + b_{mn}^l b_{mv}^{*\lambda})$$
$$+ D_{mnv}(b_{mn}^l a_{mv}^{*\lambda} + a_{mn}^l b_{mv}^{*\lambda})) \tag{48}$$

$$C_{mnv} = (-1)^{\mu} \frac{(n+m)!}{(n-m)!} \sum_{p=|n-v|}^{n+v} i^p (\cos\theta_{\lambda l})^p [n(n+1)$$
$$+ v(v+1) - p(p+1)] \frac{(p-m+\mu)!}{(p+m-\mu)!}$$
$$\times a(m,n,-\mu,v,p) j_p(kd_{\lambda l}) \tag{49}$$

$$D(m,n,\mu,v) = kd_{\lambda l} m(-1)^{m+n} i^{n+v} \frac{(n+m)!}{(n-m)!} \sum_{p=|n-v|}^{n+v} a(-m,n,m,v,p)(\cos\theta_{\lambda l})^{p+1} i^{p+1} j_p(kd_{\lambda l}) \quad (50)$$

where $\theta_{\lambda l} = \pi$ if $\theta_{l\lambda} = 0$ and $\cos\theta_{\lambda l}$ takes 1 or -1. Note that for this situation, the coefficients in Eq. (8) will not take the simple form of $m = \pm 1$, except when all the particles are aligned up along the same direction as the traveling direction of the electromagnetic waves. When this is not the case, a full expansion of (p_{mn}, q_{mn}) including all m terms is required. For a linearly polarized income wave with $\gamma = \pi$, one can show that the coefficients take the following forms,

$$\begin{aligned} p_{l,m} &= -E_0 i^{l+1} \frac{2l+1}{l(l+1)} \frac{(l-m)!}{(l+m)!} \tau_{lm}(\cos\theta_d); \\ q_{l,m} &= -E_0 i^{l+1} \frac{2l+1}{l(l+1)} \frac{(l-m)!}{(l+m)!} \pi_{lm}(\cos\theta_d) \end{aligned} \quad (51)$$

where θ_d is the angle between the wave vector and the z-axis. It is a simple matter to show that Eq. (51) reduces to Eq. (8) when the wave travels along the z-axis, i.e., $\theta_d = 0$.

It is worth noting that, for independent scattering, the term containing ($kd_{\lambda l}$) in Eq. (48) vanishes when all terms of m are summed up.

After the above derivation was developed, it was found that an expression for the total scattering cross-section of an aggregate, appearing in different form with multiple indices, also was derived using an entirely different approach in (Mackowski 1994). The indices in the expression (Mackowski 1994), however, were misplaced and thus the direct use of the expression would be difficult. To correct these misplaced indices, one may need to go through the derivations again, which can be rather complex.

Mathematically, Eq. (41) is simpler to use and requires fewer terms, while Eq. (16) needs repeated use of $A_{\mu v}^{mn}$ and $B_{\mu v}^{mn}$to translate each sphere to a common reference, and thus considerably more terms. Comparative computational tests show that Eq. (41) converges much faster and is more stable than Eq. (16). For example, for a well-studied example of two same solid dielectric spheres of $m = 1.5$ placed in a plane electromagnetic field (Olaofe 1974), Eq. (16) required 10 terms to converge to the correct result of the total scattering coefficient $= 1.81506\pi a^2$, while Eq. (41) needed only 5 terms, for the particle separation $\delta = 3$, where $\delta =$ distance between the two particles divided by the particle radius and size parameter $(2\pi a/\lambda) = 0.5$. Moreover, detailed analysis shows that for this case, with two terms, Eq. (41) yields a result that differs from the correct result by <1.0%. With two terms, however, Eq. (16) gives a result with an error of 42%. For the same case but with $\delta = 10$, Eq. (41) again needed 5 terms to converge to the correct result, while Eq. (16) required 30 terms.

The above formulas can be used to calculate the total scattering coefficients for virtually any configuration of a nanoparticle aggregate. The particles can be bare solid spheres and spheres with single- or multi-layered coatings, the only difference being the use of a_{mn} and b_{mn} for an isolated such particle.

To further test its accuracy, Eq. (41) is applied to calculate the spectra of a nanoshell dimer for which experimental measurements became available recently (Lassiter et al. 2008). The gold nanoshell consists of a silica core with a gold shell (or SiO_2@Au core/shell). The coefficients of α and β are taken from (Liu 2009),

$$\begin{aligned} \alpha^i &= \frac{m_2^i \psi_l(y^i)[\psi_l'(m_2^i y^i) - B_l \chi_l'(m_2^i y^i)] - \psi_l'(y^i)[\psi_l(m_2^i y^i) - B_l \chi_l(m_2^i y^i)]}{m_2^i \xi_l(y^i)[\psi_l'(m_2^i y^i) - B_l \chi_l'(m_2^i y^i)] - \xi_l'(y^i)[\psi_l(m_2^i y^i) - B_l \chi_l(m_2^i y^i)]}; \\ \beta^i &= \frac{\psi_l(y^i)[\psi_l'(m_2^i y^i) - A_l \chi_l'(m_2^i y^i)] - m_2^i \psi_l'(y^i)[\psi_l(m_2^i y^i) - A_l \chi_l(m_2^i y^i)]}{\xi_l(y^i)[\psi_l'(m_2^i y^i) - A_l \chi_l'(m_2^i y^i)] - m_2^i \xi_l'(y^i)[\psi_l(m_2^i y^i) - A_l \chi_l(m_2^i y^i)]} \end{aligned} \quad (52)$$

$$A_l = \frac{m_2^i \psi_l(m_2^i x^i)\psi_l'(m_1^i x^i) - m_1^i \psi_l'(m_2^i x^i)\psi_l(m_1^i x^i)}{m_2^i \chi_l(m_2^i x^i)\psi_l'(m_1^i x^i) - m_1^i \chi_l'(m_2^i x^i)\psi_l(m_1^i x^i)};$$
$$B_l = \frac{m_2^i \psi_l(m_1^i x^i)\psi_l'(m_2^i x^i) - m_1^i \psi_l'(m_1^i x^i)\psi_l(m_2^i x^i)}{m_2^i \chi_l'(m_2^i x^i)\psi_l(m_1^i x^i) - m_1^i \chi_l(m_2^i x^i)\psi_l'(m_1^i x^i)} \tag{53}$$

where subscripts 1 and 2 refer to the core and the shell, respectively, and $y^i = k^i b^i$, b^i being the outer radius of the ith nanoshell. Alternatively, these coefficients can also be obtained using the recursive algorithm (Liu and Li 2011). The material properties (Johnson and Christy 1972) with allowance for nanoscale effects (Liu and Li 2011; Okamoto 2001) were used. The results obtained from Eq. (41) (and then Eq. (45) for total scattering coefficients) are given in Fig. 2, where excellent agreement is seen between the measurements and the calculations, further validating Eq. (41). Note that both the local surface plasma resonance of the nanoshell dimer and the spectral line width are predicted very well.

We next consider another but more complicated case where a linear chain of various gold nanoshells is placed in a plane electromagnetic wave. The results for the chain of up to 10 nanoshells are shown in Fig. 3. A significant red-shift of the collective surface resonance is observed as the number of the nanoshells in the chain increases. Similar phenomenon has been reported for solid particle chains (Rechberger et al. 2003). Also, the line width of the spectra broadens

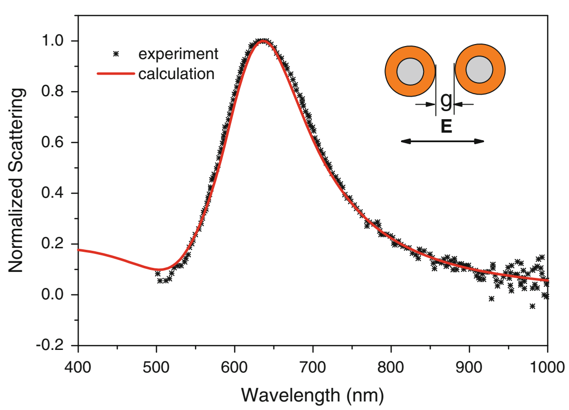

Fig. 2 Comparison of calculated and measured scattering spectrum of a nanoshell dimer ($Q_{sca} = C_{sca}/\pi a^2$, a = the outer radius of a nanoshell). The silica core and gold shell have dimensions of 42 nm radius and 17 nm-thick, and the interparticle separation between two nanoshells is 20 nm, surrounded by air. The refractive indexes of air and silica are 1 and 1.45, respectively, independent of wavelength

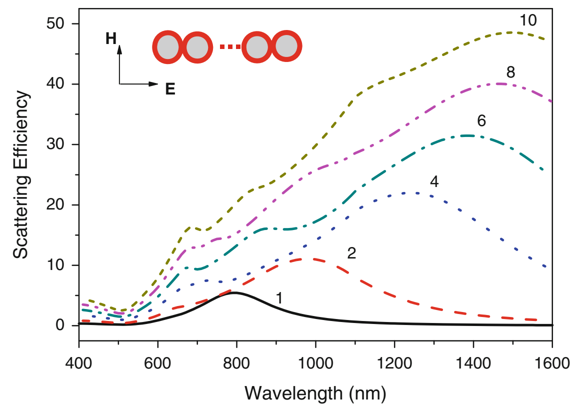

Fig. 3 Scattering efficiency of nanoshell chains. The gold nanoshells are all the same, with 60-nm radius silica core and 12 nm-thick gold shell; the gap between particles is zero. The medium is water with refractive index = 1.33. The *labels* near the curves indicate the number of particles in the chain

with more nanoshells. This is also observed in recent experiments in our laboratories (Liu et al. 2011). With Eq. (41), the following reasoning is constructed to explain the red-shift phenomenon. Near the resonance, electrons in the metal shell of two adjacent nanoshells oscillate out-of-phase, thereby canceling the effects of each other. This canceling effect, or damping effect, is illustrated by the opposite signs of leading order terms associated with the coefficients C_{mnv} and D_{mnv}. As a consequence, electrons in a nanoshell experience a damping force induced by the motion of electrons of its neighbor. This produces a level shift of the resonance peak toward the right (or red-shift), and a broadened line breadth. Put in another way, the net effect of damping force makes the two outer nanoshells at the ends of the chain mainly responsible for plasma resonance, similar to a metal-coated nanorod with its longitudinal axis in parallel with polarization. This physical picture is further supported by the calculations of linear chains of nanoshells oriented in two other directions, which both show much smaller resonance shifts. The inter-particle interaction was conjectured to affect the restoring force for electrons as the mechanism for level-shifts (Rechberger et al. 2003). Eq. (41) suggests that it is more appropriate to interpret the interaction as a damping force, as the latter also results in line width broadening (Jackson 1976; Messiah 1981). Simulations were made also for many different particle configurations and orientations. Numerical data and experiments show that Eq. (41) converges fast and is stable and that the

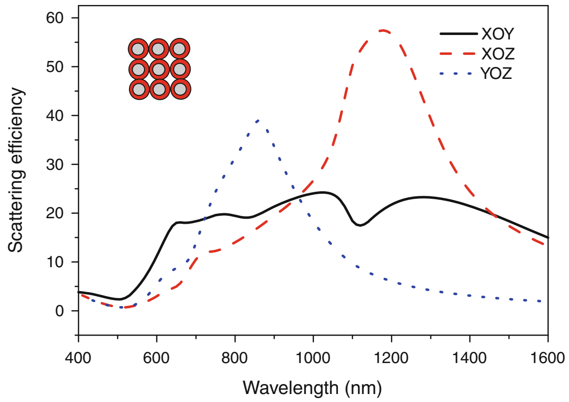

Fig. 4 Scattering efficiency of an array of nanoshells. The *solid*, *dashed*, and *dotted curves* correspond to the collective scattering efficiency of the arrays arranged in XOY, XOZ, and YOZ plane, respectively. Other conditions are the same as in Fig. 3

resonance behavior of these arrangements can be interpreted from the combined effects of the linear chains. One of these cases is shown in Fig. 4, which depicts the spectra of the total scattering coefficient of 9 nanoshells arranged to form a regular square.

Concluding remarks

A new approach has been presented to derive a general analytical expression of the total scattering coefficient for an aggregate of multiple nanoparticles. A simplified expression is obtained by following the same procedure for a linear chain of particles. These expressions are useful in clearly elucidating the nature of the interactive coupling between nanoparticles during multiple scattering of electromagnetic waves. Calculated results are in excellent agreement with existing measurements for a nanoshell dimer. For a linear chain of particles with the chain axis parallel to the polarization direction of the electric field, a significant red-shift of resonance peaks occurs mainly because electrons in metal shells of two adjacent particles oscillate out-of-phase to cancel each other's radiation effects. The damping effect resulting from the out-of-phase oscillation also broadens the line width of the scattering spectra. Multi-scattering by more complex configurations of nanoshell aggregates may be attributed to the combined effects of linear chains of nanoshells.

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