Spectral Responses by Chains of Subwavelength-Size Metallic Spheres in Terahertz (THz) Region

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Abstract: The extinction spectra for chains of subwavelength-sized metal spheres responding to incident terahertz plane waves are calculated in 0.1-2.5*THz* frequency range using a generalized Mie theory. It is found that the coherent coupling between the spheres varies as the number N of spheres in the ensemble increases and finally approaches a steady response, which manifests that the collective excitation of the spheres only exists within a finite spatial scope. Therefore, the spectral responses of the systems strongly depend on the polarization of incident waves, diameter and interspacing of spheres while the detailed conductive properties of metals only play a minor role. Especially, the interaction of the L-mode incident field whose electric field parallel to the array of spheres, with the ensemble, is much stronger than that of the T-mode and shows complicated resonant features.

Key words: terahertz, metamaterials, metallic spheres, extinction spectrum

1. Introduction

The interaction of subwavelength-size metallic structures with electromagnetic (EM) waves has attracted great research interest recently stimulated by the advancement of material fabrication and optical science. In many cases, the surface plasmon polaritons (SPPs) excited in the systems play a key role and many unusual phenomena were observed, such as enhanced transmission of a small hole or hole array [1, 2], effective wave guiding by metal wires [3], manipulation of light propagation in the split ring resonators and the so-called left-hand material with negative refractive index [4, 5], excitation and propagation of SPP waves on the corrugated metal surface and in random subwavelength structures [6], and near-field coupling between metallic nanoparticles [7], etc. Though there is virtually no dispute in the fundamental physical picture on these phenomena and it is true that all those observations are related to the resonant excitation of charge density waves and associated evanescent EM waves, the detailed elucidation and explanation to a specific phenomenon could still show some divarication [8, 9, 10, 11]. Therefore, further exploration is required in order to understand the physics and developing device applications based on the functional metastructures. In this paper, utilizing a generalized Mie theory, a multiple-scattering method based on the spherical-wave expansion of the electromagnetic field, we investigate the spectral responses of 1-D chains of metal spheres to the incident terahertz (THz) plane waves and the dependence on the metal conductive properties, ensemble structure geometries and the polarization of the incident THz waves. In the calculated extinction spectra, distinct resonant features are observed for the incident THz field with an electric vector pointing along the array direction, but the resonant features cannot be directly connected to the periodicity of the sphere chains, or surface plasmon modes.

2. Theory and method

The electromagnetic response of bulk metals is commonly described by the collective oscillations of electrons, i.e., Drude permittivity $\varepsilon = \varepsilon_r + i\varepsilon_i = 1 - \omega_p^2/(\omega^2 + i\omega\gamma)$, where ω_p and γ are the plasma frequency and damping frequency respectively. The typical particle diameter in our simulation is $100\mu m$, which is about three orders of magnitude larger than the skin depth δ. Thus we take the Drude model to describe the dielectric function of the metallic spheres. In our study, the spectral responses of the ensembles of spheres are calculated using the modal expansion method based on the generalized Mie theory, developed in a series of papers by Bruning and Lo [12], Gérardy and Ausloos [13], and Fuller [14]. Under this theoretical frame, it's convenient to calculate the extinction cross section of wave scattering, which counts the sum of the absorption and scattering losses, and the results can be easily compared to the experiment measurements.

Considering an incident plane wave scattered by a single spherical particle, the incident and scattered fields can be expanded in terms of vector spherical harmonics,

$$
\begin{aligned}
\mathbf{E}_{inc}(\mathbf{r}) &= \sum_{q=1}^{\infty}\sum_{p=-q}^{q} [a_{qp}\mathbf{m}_{qp1}(\mathbf{r}) + \boldsymbol{b}_{qp}\mathbf{n}_{qp1}(\mathbf{r})],\\
\mathbf{H}_{inc}(\mathbf{r}) &= \sum_{q=1}^{\infty}\sum_{p=-q}^{q} [b_{qp}\mathbf{m}_{qp1}(\mathbf{r}) + a_{qp}\mathbf{n}_{qp1}(\mathbf{r})],\\
\mathbf{E}_{sca}(\mathbf{r}) &= \sum_{q=1}^{\infty}\sum_{p=-q}^{q} [c_{qp}\mathbf{m}_{qp3}(\mathbf{r}) + d_{qp}\mathbf{n}_{qp3}(\mathbf{r})],\\
\mathbf{H}_{sca}(\mathbf{r}) &= \sum_{q=1}^{\infty}\sum_{p=-q}^{q} [d_{qp}\mathbf{m}_{qp3}(\mathbf{r}) + c_{qp}\mathbf{n}_{qp3}(\mathbf{r})], \qquad (1)
\end{aligned}
$$

where q is the multipolar order and that the term with q = 1, 2, 3,… is called the dipole mode, quadrupole mode, octupole mode, et al, respectively. The Bessel spherical functions, $\boldsymbol{m}_{qp1}$, $\boldsymbol{n}_{qp1}$, and the first kind Hankel functions, $\boldsymbol{m}_{qp3}$, $\boldsymbol{n}_{qp3}$ are chosen to meet the required asymptotic behavior at infinity to facilitate the implement of the boundary conditions. The coefficient set $\{a_{qp}, b_{qp}\}$ describes the incident waves and is given by the orthogonal relation. The unknown coefficients $\{c_{qp}, d_{qp}\}$ representing the scatter waves by the sphere will be determined by applying the boundary conditions of electromagnetic fields on the surface of the sphere, and simple relations are produced,

$$
\begin{aligned}
c_{qp} &= \Gamma_q a_{qp}\\
d_{qp} &= \Delta_q b_{qp} \qquad (2)
\end{aligned}
$$

Here $\mathit{\Gamma}_q$ and $\mathit{\Delta}_q$ are the 2^q polar electric and magnetic susceptibilities of the sphere, respectively, defined in Mie scattering theory. They are related to dielectric properties and the size of the sphere.

When a plane wave is incident onto an ensemble consisting of N spherical particles, each individual particle is polarized not only by the incident field $\boldsymbol{E}_{inc}$, $\boldsymbol{H}_{inc}$, but also by the scattered fields from other particles. Therefore, the contributions to the total scattering fields from the ith sphere, i.e., $\boldsymbol{E}^{(i)}_{sca}(\boldsymbol{r})$ and $\boldsymbol{H}^{(i)}_{sca}(\boldsymbol{r})$, take the same expressions in the local coordinate system as in Eq.(1) but the expansion coefficients $c^{(i)}_{qp}$ and $d^{(i)}_{qp}$ now should include both the response to the direct incident field and the coupling with other spheres. After expanding $\boldsymbol{m}^{(i)}_{qp3}$ and $\boldsymbol{n}^{(i)}_{qp3}$ in terms of $\boldsymbol{m}^{(j)}_{qp1}$, $\boldsymbol{n}^{(j)}_{qp1}$ and applying the boundary conditions on each sphere's surface, we get the equation set about $c^{(i)}_{qp}$ and $d^{(i)}_{qp}$ as follows,

$$c_{qp}^{(i)} = \Gamma_q^{(i)}\{a_{qp}^{(i)} + \sum_{j\neq i}^{N}\sum_{l,m}[c_{lm}^{(j)}T_{lmqp}^{(i,j)} + d_{lm}^{(j)}L_{lmqp}^{(i,j)}]\},$$

$$c_{qp}^{(i)} = \Gamma_q^{(i)}\{a_{qp}^{(i)} + \sum_{j\neq i}^{N}\sum_{l,m}[c_{lm}^{(j)}T_{lmqp}^{(i,j)} + d_{lm}^{(j)}L_{lmqp}^{(i,j)}]\}, \qquad (3)$$

where $T_{lmqp}^{(i,j)}$ and $L_{lmqp}^{(i,j)}$ represent the propagation factors of the scattered electromagnetic fields from the jth sphere to ith sphere and contain the information of the spatial distribution of the spheres. Obviously this equation set is self-consistent and takes into account the coupling between the spheres. In practice, it is found that the contributions by higher multipolar modes in Eq.(1) become negligible when the order q reaches a curtain value. Therefore, in our numerical calculation the upper limit of q is restricted to a truncated value M.

The average extinction cross section of a sphere in the ensemble can be obtained as

$$Q_{ext} = \frac{1}{N\pi k^2R^2}\sum_{j=1}^{N}\sum_{q,p}\mathrm{Re}[a_{qp}^{(j)*}c_{qp}^{(j)} + b_{qp}^{(j)*}d_{qp}^{(j)}], \qquad (4)$$

where Re functions to take the real part and the asterisk denotes the complex conjugate, and Q_{ext} is measured in the unit of the geometric projection of a sphere. This quantity accommodates both the absorption and scattering effects and characterizes the responses measured in the far-field of the system to the incident field, which depends on the material properties and, especially, on the structure geometry of the ensemble.

In our calculations, linear chains of Al spheres with different particle number, the particle size and interspacing are investigated. The material parameters of aluminium are taken as ω_p = $2.243\times10^{16}Hz$ and γ = $1.22\times10^{14}Hz$ [15], which are used to produce the dielectric function by Drude model.

3. Results and Discussions

First, we study the effects of the particle numbers in the ensemble on the spectral responses. This kind of dependence results from the coupling of electric polarizations between particles through collective near-field interaction. It is clear that the different polarization of the incident field will change the induced polarization in the 1-D linear chains of metallic spheres. Two spatial cases are considered here, i.e. the incident field polarized parallel and perpendicular to the spheres array, which correspond to the longitudinal mode (L-mode) and the transverse mode (T-mode) excitation, respectively. Fig. 1(a) and Fig. 1(b) depict calculated extinction spectra versus the number of spheres N in the T-mode and L-mode respectively. Here the spheres have the same radius of $R=50m$ and the spacing of adjacent spheres is $d=2R+0.1R$. The maximum multipolar order in the calculations is taken as $M=5$.

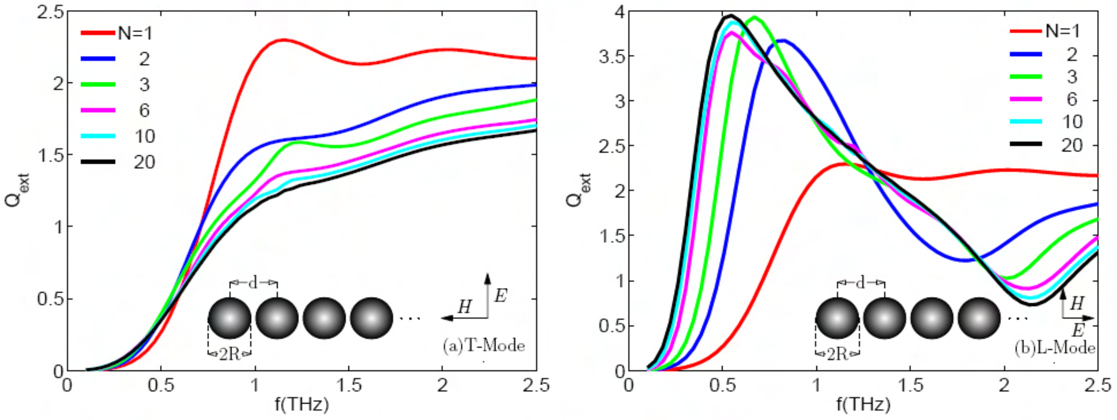

Fig.1 The calculated extinction spectra of *T*-mode(*a*) and *L*-mode(*b*) for chains with different numbers of spheres as $N = 1, 2, 3, 6, 10, 20$. $R = 50^{1}m$ and $d = 2R + 0.1R$. The summation in Eq(1) for the multipolar order is performed up to M=5.

In all the cases, the extinction cross sections increase rapidly at the lower end of the frequency spectra till reaching a maximum, which is a manifestation of the frequency dependent diffraction of the ensemble to the incident waves. However, as the number of spheres increases, the Q_{ext} for the *T*-mode excitation declines monotonously and have a flat response while the *L*-mode causes higher extinction and shows complicated resonant structures. An important observation is that the extinction spectra for both *T*- and *L*-mode incident fields continue to change as the spheres' number *N* increases so as to reach steady responses for *N* approaching to 20 finally. Therefore, the collective polarization of the spheres in the ensemble plays a key role in the responses of the ensemble, and the inter-sphere coupling only keeps within a limited spatial scale, which covers about 20 spheres in the current configuration. This dimension is about 7 times free-space wavelength at 1*THz* and should be within the lower limit of the coherent coupling range between spheres in the ensemble. It is noticeable in Fig. 1(b) that there is a nearly constant extinction cross-section value Q_{ext} at about 1.26*THz* independent of the spheres' number and, at this frequency, the system's behavior is less affected by the collective excitation of the ensemble. In a separated study on the electric field propagation in this system under a local excitation of only one sphere in the edge of the chain, we find that there exist some low dissipation surface waveguide modes, especially at frequency near $c=2d$ and $c=d$ which can be related to the excitation of surface plasmons. But this feature was not observed in the data of this paper. The reason might be due to the different excitation condition and that the far-field spectra calculated here cannot reflect the detailed resonant coupling and decoupling between the free-space EM waves and evanescent fields. Finally, we would like to point out that the chains of spheres effectively scatter the incident THz waves except at frequency about 2.15*THz* for *L*- mode, where Q_{ext} <1 for large spheres' number. Because the *T*-mode dose not induce strong coupling between spheres and show little spectral features and we just present and discuss the data of the *L*-mode hereafter.

The dependence on the sphere size for chains with N =20 is shown in Fig.2(a) for a constant ratio d/R =2.1. Under this arrangement, the separation of spheres was kept much smaller than their size and, therefore, the strong near field coupling between the spheres is maintained as the radius and spacing increase simultaneously. The noticeable feature here is that both the extinction and transmission maxima shift to lower frequency dramatically when the sphere size R increases from 20μm to 50μm. Carefully inspecting the shift of the extinction maximum, we also have found that the peak frequency is inversely proportional to the sphere size. This feature can be understood by the scale invariance of Maxwell equation if the frequency dependence of the dielectric function is ignored in the frequency range under investigation. As the sphere size R is maintained constant and the inter-particle spacing d/R changes independently from 2.1 to 3.3, the extinction maximum shifts to higher frequency (Fig. 2(b)) and, meanwhile, the peak value drops quickly. Considering the relative weight of the contributions from the size dependent diffraction by a single sphere and the near-field coupling effects between spheres by the near field interaction, it is expected that when $d >> R$, the spectral responses of the spheres' chain will tend toward that of an isolated sphere as shown in Fig.1. Therefore, as the inter-particle spacing increases, the reduced coupling between spheres makes the system's responses approach the situation of the isolated sphere and, as a result, the extinction maximum becomes smaller and shifts to the higher frequency. The transmission maximum moves to lower frequency as the sphere separation increases, which is correlated to the change of the reciprocal lattice vectors but no conclusive or special relation is found.

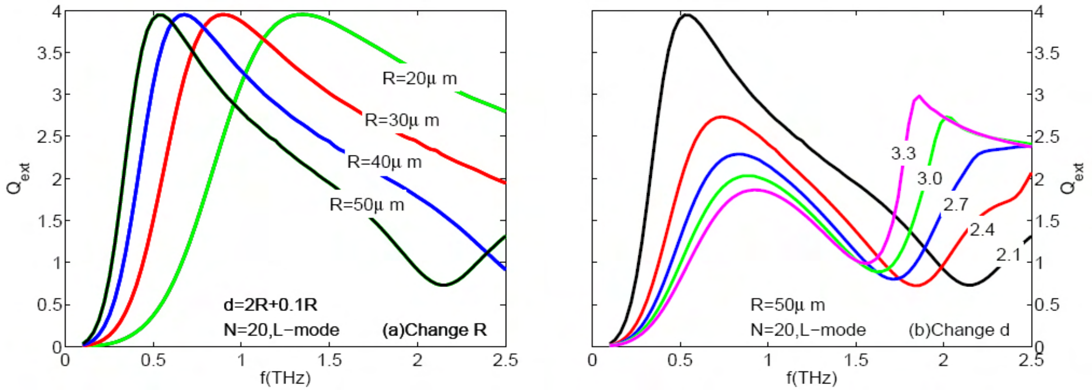

Fig.2 (a) The spectra at d = 2.1R for the different size R of particles and (b) the spectra at R = 50μm for the interspacing d=R = 2.1, 2.4, 2.7, 3.0, and 3.3.

In our calculations, we have found that the extinction spectra have little relation with the detailed dielectric properties of metals and we get almost the same results after replacing Al particles by *Au* (ω_p=1.366×10^{16}*Hz*, γ =4.071×10^{13}*Hz*) and *Cu* (ω =1.203×10^{16}*Hz*, γ = 5.240×10^{13}*Hz*) [15], which is quite different from the observations in optical region [2, 16]. In THz region, the drastic increase in dielectric constants of metals makes them behave like an ideal metal somewhat and the detailed structure features play a dominant role in the ensemble's responses to the incident EM field. This property provides an extra flexibility in choosing metal materials for functional devices based on the near-field interaction in the THz region.

In the frame of the generalized Mie theory used here, the precision of calculations depends on the maximum order *M* of the multipolar modes. As an example, the extinction spectra of Al bispheres with R=50μm and d =2R +0.1R versus *M* are shown in Fig.3. Choosing M=5 is adequate for the system investigated here. For larger particles or a shorter spacing between spheres, higher multipolar modes should be considered [17].

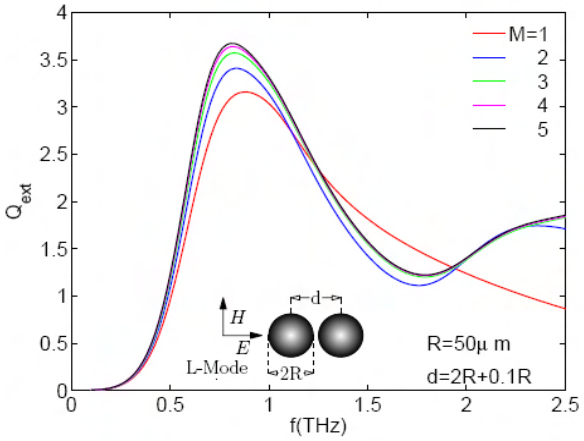

Fig.3 The extinction spectra of Al bispheres with the maximum multipolar mode M = 1, 2, 3, 4, 5 taken in calculations. R = 50μm and d = 2R + 0.1R

4. Conclusions

In summary, the extinction spectra for chains of metal spheres in 0.1-2.5*THz* range are calculated and analyzed using a generalized Mie theory. Because the frequency of the incident EM field is well below the bulk plasmon frequency of metals, the spectral responses of the systems depend much more on the polarization of incident waves and the structure geometry of the ensemble, while the detailed conductive properties of metals only play a minor role. The spectral features show that the near field coupling between metallic spheres dominates the system's responses and the collective excitation of the spheres only exists within a finite spatial scope. Our calculations demonstrate that the detailed spectral responses of the ensemble can be tailored greatly by changing its structural parameters with a considerable flexibility, which is very useful in functional device applications.

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