Journal of Zhejiang University SCIENCE A
ISSN 1009-3095
http://www.zju.edu.cn/jzus
E-mail: jzus@zju.edu.cn

A resonator mode in linear arrays of silver spheres and cylinders

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Received Sept. 5, 2005; revision accepted Oct. 10, 2005

Abstract: A transversal mode with zero group velocity and non-zero phase velocity that can exist in chains of silver nano-spheres in the optical frequency range was theoretically studied. It is shown that the external source radiating a narrow-band non-monochromatic signal can excite in the chain a mixture of standing and slowly travelling waves. The standing wave component (named as resonator mode) is strongly dominating. The physical reason of such a regime is a sign-varying distribution of power flux over the cross section of the chain. This situation is similar to the scenario of the propagation of a wave along the boundary between the right-handed and left-handed media where the spatial distribution of the light intensity is vortex. However, in the present case there is no boundary between media and the boundary between the positive and negative power fluxes is a cylindric tube in free space whose axis is the axis of the chain.

Key words: Resonator mode, Linear arrays, Plasmon particle
doi:10.1631/jzus.2006.A0029 **Document code:** A **CLC number:** O441; TN75

INTRODUCTION

The waveguide properties of linear arrays (chains) of silver nano-particles at optical frequencies are well known (Krenn *et al*., 1997; 1999; Tretyakov and Viitanen, 2000; Girard *et al*., 1994; Weber and Ford, 2004). Silver spheres and ellipsoids can support forward or backward waves along the chain depending on their polarization state and the eigenfrequency (Krenn *et al*., 1999; Tretyakov and Viitanen, 2000; Girard *et al*., 1994; Weber and Ford, 2004). At comparatively low frequencies (the wavelength λ in host medium is much larger than the period) the longitudinal polarization of silver nano-spheres (along the chain axis) corresponds to a forward wave. The transverse polarization of spheres can correspond to a backward or a forward regime depending on the relations between the sphere diameter d, array period a and wavelength λ. The waveguide properties of silver nano-cylinders are weakly studied. To our knowledge, the only case of longitudinal polarization was considered in the literature and the forward eigenwave was obtained. Cylinders (silver nano-wires) are easier to prepare and their waveguide properties deserve no less attention than the waveguide properties of nano-spheres.

A complete analytical model of eigenwaves in a linear chain of silver spheres was reported (Viitanen *et al*., 2005). A similar model for an array of silver nano-wires has been also developed. In the present paper we concentrate on an interesting effect corresponding to a special eigenfrequency that appears in spheres and cylinders when their diameter becomes approximately 1/10 that of λ at the plasmon resonance. The wave excited in the array by a finite source turns out to be a mixture of a standing wave and a travelling (backward) wave. The standing wave is not the Bragg mode and does not occur at the edge of the Brillouin zone. In the dispersion diagram it corresponds to the zero group velocity and nonzero phase velocity. This wave can be named as a resonator mode.

DISPERSION EQUATIONS

Let a z-polarized sphere centered at $x=a/2$ be chosen as a reference particle and z-directed dipole moment p is attributed to it. All other spheres are modelled as z-directed dipoles p_n whose phase shift with respect to the reference one is determined by propagation constant q: $p_n=p\exp(-\mathrm{j}qna)$. The same procedure is implied for cylinders, where p denotes the polarization per unit length (PUL) of a cylinder stretched along y.

Neglecting the losses in silver (in order to obtain a real-value dispersion equation), we can write the expression for the inverse polarizability of a sphere or a cylinder (polarized across y) in the following (Weber and Ford, 2004; Sareni, 1996):

$$\frac{1}{\alpha}=\frac{\varepsilon_\mathrm{s}+2\varepsilon}{3\varepsilon_0\varepsilon V(\varepsilon_\mathrm{s}-\varepsilon)}+\mathrm{j}\frac{\kappa^3}{6\pi\varepsilon_0\varepsilon},$$
$$\frac{1}{\alpha}=\frac{\varepsilon_\mathrm{s}+2\varepsilon}{2\varepsilon_0\varepsilon S(\varepsilon_\mathrm{s}-\varepsilon)}+\mathrm{j}\frac{\kappa^2}{8\pi\varepsilon_0\varepsilon}. \tag{1}$$

Here $V=\pi d^3/6$ is the sphere volume, $S=\pi r^2=\pi d^2/4$ is the cylinder cross section, $\kappa=\omega\sqrt{\varepsilon_0\mu_0\varepsilon}$ is the host medium wave number and ε is the matrix permittivity. The real part of Eq.(1) corresponds to the commonly known static polarizability of a dielectric sphere or a cylinder, and the imaginary part describes the re-radiation of the polarized dipole or a dipole line, respectively. The local field $\boldsymbol{E}^{(\mathrm{loc})}$ (z-component) acting on the reference dipole (or dipole line in the case of cylinders) is produced by all the other dipoles and can be expressed through the reference dipole moment $p=\alpha\boldsymbol{E}^{(\mathrm{loc})}$ in terms of the interaction constant of the array: $\boldsymbol{E}^{(\mathrm{loc})}=C(\kappa, q, a)p$, which depends on the matrix wave number k, the propagation constant q and the array period a. The dispersion equation $1/\alpha=C$ transits to the real equation $\mathrm{Re}(1/\alpha)=\mathrm{Re}(C)$ since outside the light cone the imaginary parts of the interaction constant and these of the inverse polarizability cancel out. The interaction constant of the chain of transversally polarized dipoles in a homogeneous matrix of permittivity ε was obtained in (Viitanen *et al*., 2005) in the form appropriate for fast calculations, with the dispersion equation being written as follows:

$$-\frac{\varepsilon_\mathrm{s}+2\varepsilon}{3V(\varepsilon_\mathrm{s}-\varepsilon)}=\frac{\boldsymbol{k}^2}{4a\pi}\ln(2|\cos qa-\cos \boldsymbol{k}a|)$$
$$=\frac{1}{2\pi}\sum_{n=1}^{+\infty}\left[\frac{\boldsymbol{k}\sin n\boldsymbol{k}a}{(na)^2}+\frac{\cos n\boldsymbol{k}a}{(na)^3}\right]\cos nqa \tag{2}$$

To find the interaction constant for a linear array of parallel cylinders, we used the known formulae for the field of a dipole line (Felsen and Marcuvitz, 1994) and some properties of Bessel functions. The dispersion equation takes the form:

$$\frac{d}{\boldsymbol{k}r^2}\frac{\varepsilon_0+\varepsilon}{\varepsilon_0-\varepsilon}=\ln\left(\frac{\boldsymbol{k}d\gamma}{4\pi}\right)+\frac{\pi}{\sqrt{(qa)^2-(\boldsymbol{k}a)^2}}$$
$$+\frac{1}{2\pi}\sum_{n=1}^{+\infty}\left[1\Big/\sqrt{\left(n+\frac{qa}{2\pi}\right)^2-\left(\frac{\boldsymbol{k}a}{2\pi}\right)}-\frac{1}{n}\right]$$
$$+\frac{1}{2}\sum_{n=1}^{+\infty}\left[1\Big/\sqrt{\left(n-\frac{qa}{2\pi}\right)^2-\left(\frac{\boldsymbol{k}a}{2\pi}\right)}-\frac{1}{n}\right]$$
$$+\pi\sum_{n=1}^{+\infty}\frac{Y_1(\boldsymbol{k}na)\cos qna}{n}. \tag{3}$$

Silver's permittivity to be substituted into Eqs.(2) and (3) is as follows: $\varepsilon_\mathrm{s}=1-\omega_\mathrm{p}^2/\omega^2$ where the plasma frequency ω_p corresponds to a wavelength of 150~300 nm depending on the particle radius r (Kreibig, 1970).

The solution of the dispersion Eqs.(2) and (3) for the following parameters: $a=1.5d=75$ nm, $\lambda_\mathrm{p}=272$ nm, $\varepsilon=1$ are shown in Fig.1. Fig.1a corresponds to the array of spheres, Fig.1b is for nano-wires. The zero group velocity (resonator mode) corresponds to $\omega_\mathrm{r}=0.999\omega_0$, $q_\mathrm{r}=0.401\pi/a$ in the case of spheres and to $\omega_\mathrm{r}=1.078\omega_0$, $q_\mathrm{r}=0.545\pi/a$ in the case of nano-wires. Here ω_0 corresponds to the plasmon resonance of nano-particles: $\omega_0=\omega_\mathrm{p}/\sqrt{3}$ and $\omega_0=\omega_\mathrm{p}/\sqrt{2}$ for spheres and cylinders respectively.

EXCITATION OF PLASMON ARRAYS

We studied the excitation of both arrays of spheres and cylinders by a finite source, namely by a z-polarized dipole line located on the y-axis with dipole moment PUL $P(t)$. Let the source temporal dependence be $P(t)$ so that the dipole line radiates a pulse with very narrow frequency band $\Delta\omega$ containing the frequency of the resonator mode ω_r. The problem was solved in terms of Fourier harmonics. At every frequency the field of the source was expanded into spatial spectrum defined as:

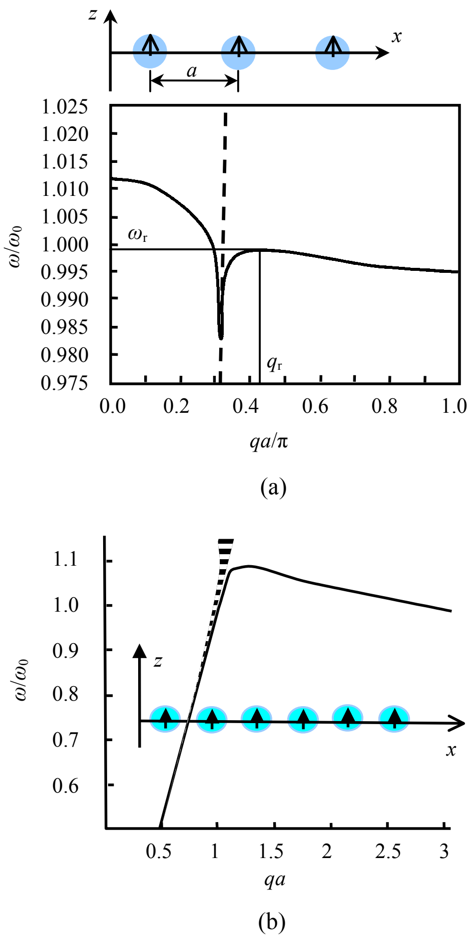

Fig.1 Dispersion diagrams. (a) Spheres. Light line is dashed; (b) Cylinders. Leaky mode is shown as a dotted line of variable thickness

$$f(\boldsymbol{k},k_x,z)\equiv\frac{1}{2\pi}\int_{-\infty}^{\infty}f(\boldsymbol{k},x,z)\mathrm{e}^{-\mathrm{j}k_x x}\mathrm{d}x.$$

The z-component of the external field has the spatial spectrum

$$E_z^{\mathrm{e}}(\boldsymbol{k},k_x,z)=\frac{k_x^2P(\boldsymbol{k})\mathrm{e}^{\mathrm{j}\sqrt{\boldsymbol{k}^2-k_x^2}|z|}}{\left(2\varepsilon_0\sqrt{\boldsymbol{k}^2-k_x^2}\right)}\ . \tag{4}$$

The dipole moment of a sphere or that of a cylinder (PUL) corresponding to a spatial harmonic $k_x=q$ is given by $p=\alpha[E_z^{\mathrm{s}}(\boldsymbol{k},q,0)+C(\boldsymbol{k},q)p]$. For the lossless structure it can be rewritten as:

$$p(\boldsymbol{k},q)=E_z^{\mathrm{s}}(\boldsymbol{k},q,0)\mathrm{e}^{-\mathrm{j}qa/2}\Big/(1/\alpha-C(\boldsymbol{k},q)). \tag{5}$$

Let us introduce small losses in the silver permittivity $\varepsilon_{\mathrm{s}}=1-\omega_{\mathrm{p}}^2/\omega^2-\mathrm{j}\omega_{\mathrm{p}}^2\omega_{\mathrm{D}}/\omega^3$, where ω_{D} is the damping frequency, $\omega_{\mathrm{D}}<<\omega$. For all q except solutions of dispersion in Eq.(2) or Eq.(3), the denominator in Eq.(5) is high, and the excitation is negligible. For eigenmodes $\mathrm{Re}(1/\alpha)\approx\mathrm{Re}(C)$ and the excitation is resonant. For example, for the chain of spheres, the induced dipole moment is as follows:

$$\begin{aligned}p(\boldsymbol{k})=&\left(\mathrm{j}q^2P(\boldsymbol{k})\mathrm{e}^{-\mathrm{j}qa/2}\Big/\left(2\varepsilon_0\sqrt{\boldsymbol{k}^2-k_x^2}\right)\right)\\&\times\left(\varepsilon_0V\omega^3\big/\omega_{\mathrm{p}}^2\omega_{\mathrm{D}}\right)\times\left(\omega_{\mathrm{p}}^2\big/\omega^2+\varepsilon_{\mathrm{s}}-1\right)\\&\times\left(\omega_{\mathrm{p}}^2\big/\omega^3+\varepsilon_{\mathrm{s}}-1-\mathrm{j}\omega_{\mathrm{p}}^2\omega_{\mathrm{D}}\big/\omega^3\right).\end{aligned} \tag{6}$$

A similar formula was obtained for cylinders. At every frequency the field $E_z^{\mathrm{s}}(\boldsymbol{k},q,0)$ excites two waves: one is forward with wave number $q=q_{\mathrm{f}}$, the other is backward $q=-q_{\mathrm{b}}$, as shown in Fig.2. Really, the source is positioned at $x=0$ and at $x>0$ the group velocity of both excited eigenwaves should be directed to the right. This means that for the backward wave we should choose the negative dispersion branch. So, the total field at given $\boldsymbol{k}$ is produced by two polarizations of the chain given by Eq.(6): one corresponds to $q_{\mathrm{f}}(\boldsymbol{k})$, the other corresponds to $q_{\mathrm{b}}(\boldsymbol{k})$.

To every time harmonic of the radiated pulse, the axial Poynting vector corresponds to that which includes the contribution of both forward and backward waves and their cross products. We studied the spatial distribution of S_x using Eq.(6) and applying Floquet expansion for the components of the electric and magnetic field produced by dipoles $p(\boldsymbol{k})$. In a very narrow tube around the axis of the chain of spheres

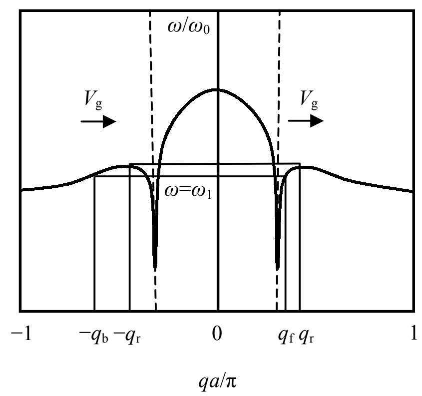

Fig.2 Two waves, forward one and backward one both with V_g>0 are excited in the right part of the chain (with respect to the source) at the frequency $\omega_1 \approx \omega_r$. At the frequency ω_r both q_f and q_b transit to q_r and $V_g \rightarrow 0$

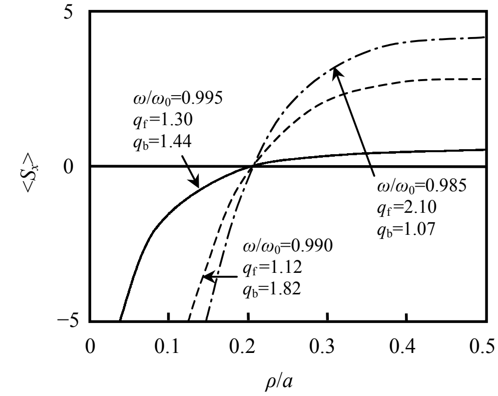

Fig.3 Power density distribution (in arbitrary units) in the vicinity of the chain axis when the two-mode regime is excited in the chain corresponding to Fig.1 at three different frequencies close to ω_r

the energy propagates backward and outside it propagates forward. The forward energy flux dominates and we obtained

$$P_x = \int_0^\infty <S_x^t> \rho \mathrm{d}\rho, \tag{7}$$

where brackets < > denote the averaging over the azimuthal angle and over the array period a. However the total power flux is much smaller than that of mode q_f or that of the mode q_b taken separately. The value ρ is a parameter of the chain that almost does not depend on the frequency. The distribution of the averaged Poynting vector around the chain axis is shown in Fig.3 for three different frequencies. When $\omega \rightarrow \omega_r$ the axial Poynting vector tends to zero. A similar result was obtained for the array of cylinders when the backward energy flux holds in a thin layer centered at the plane OXY. Applying inverse Fourier transform to the z-component $E_z(\boldsymbol{k},q(\boldsymbol{k}),0)$ of the field produced by the chain, we also obtained an approximate formula:

$$<E_z(x,t)> \approx \cos q_r x F(t) + O(\Delta\omega/\omega_r), \tag{8}$$

here brackets < > denote the averaging over the effective cross section of the guided mode, $O(\Delta\omega/\omega_r)$ is a small function of (x,t) representing the travelling wave (which is backward) and $F(t)$ is proportional to $P(t)$, i.e., represents the temporal dependence of the source. The first (dominating) term is a standing wave. The physical meaning of these results is simple: one half of the pulse is a quasi-monochromatic forward wave and the other half is a quasi-monochromatic forward wave. The presence of the travelling-wave component is due to the small non-symmetry of the dispersion branch around the point $q=q_r$.

One can split the positive power flux corresponding to the integral of the Poynting vector $<S_x^t>$ over the domain $\rho>\rho_0$ into two parts. One part is equal to the negative power flux (the integral over the domain $\rho<\rho_0$) and corresponds to the standing wave in Eq.(8). The other part, the difference of positive and negative fluxes, corresponds to the small travelling wave component in Eq.(8).

The effective group velocity of the pulse is approximately equal to the frequency band divided by the band of the wave numbers of the pulse harmonics (Fig.2). $V_g \approx \Delta\omega/\Delta q \approx \Delta\omega/2q_r$, i.e., V_g is as small as the energy flux P_x.

In practical cases such pulse whose field is close to a standing wave will be excited in a finite (due to losses) but rather long portion of the chain. Recall that the Bragg mode of an infinite periodic structure (which is formally also a resonator mode) is not supported by the chain since it locates just at the edge of the band-gap. It corresponds to the rapidly attenuating excitation of few elements of a periodic structure. And usual standing waves arise due to the reflection from some bounds.

CONCLUSION

In this paper we theoretically demonstrated the possibility to excite a standing wave in the infinite chain of plasmon particles (silver nano-spheres and nano-cylinders) in the optical range. This standing wave (resonator mode) can be excited by a source positioned on the chain axis or near it and exists together with the travelling wave which has much smaller amplitude. The frequency at which such regime can be fulfilled corresponds to the special point on the dispersion plot of the chain where the group velocity becomes zero and the phase velocity does not.

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