Reduced radiation losses in electron beam excited propagating plasmons

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Abstract: Except for heating losses in metal, propagating plasmons also suffer a lot from radiation losses. In this paper, electron beams are proposed as a way to excite higher-order, multipolar plasmons, which would otherwise not be excited by light, as a way to reduce radiation losses. Specifically, electron excited guided plasmons in a coupled nanoparticle chain and a symmetrical four-wire waveguide are separately discussed. In the coupled nanoparticle chain, the plasmon mode formed by quadrupolar polarized particles with low radiation is efficiently coupled by electron beams. Meanwhile, in the four-wire waveguide, the excited plasmons with zero momentum in the cross-section of each wire possess longer propagating distance than other higher-order plasmons.

OCIS codes: (240.6680) Surface plasmons; (250.5403) Plasmonics; (230.7370) Waveguides.

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1. Introduction

Surface plasmon polaritons (SPPs) are collective excitation of electron density waves and optical waves at the surface of metals [1]. Their extremely high band width and the ability breaking through the diffraction limit make them potential information carrier in future communication technology. Various metal structures are proposed to guide SPPs by resolving the conflict between heating losses and spatial confinement to achieve long propagation distance, including planar metal films [2], channels patterned in metal [3], metallic wedges [4], coupled nanoparticle arrays [5], closed wires [6] and metellodielctric hybrid structures [7]. And all of these designs are based on the idea of little metal penetration. However, the propagation distance of SPPs is limited not only by the heating losses in metal but also by the radiation losses into far field. For instance, the radiation losses exceed ohmic losses for small particles in particle arrays. As we know, little attention has been paid on how to reduce the radiation losses of SPPs, which mainly due to that only dipolar plasmon modes are usually excited by light in nanostructures.

Meanwhile, an electron beam is believed to be an efficient plasmon source due to the evanescent field accompanying by moving electrons [8–11]. And electron energy loss spectrum (EELS) and cathodoluminescence spectrum (CL) are used as real nanoscale plasmon mapping methods because of shorter wavelength of electrons than photons [12]. In addition, compared with light, fast electrons possess larger momentum than photons thus making higher-order, multipolar plasmons excitation possible, e.g. quadrupolar plasmons in silver nanoparticles have been mapping using an electron beam [13]. Therefore, propagating higher-order, multipolar plasmon modes which cannot be excited by light should can be coupled by electrons. More importantly, these kinds of plasmons are silent to light, in principle, could lower radiation losses due to their born light-shielding character.

Based on this idea, in this paper two common used metallic structures, a nanoparticle array and a multi-wire waveguide are examined. Specifically, guided plasmon modes generation by electron beams in the coupled nanoparticle chain and the four-wire waveguide are studied using exact electrodynamic methods, respectively. In the coupled particle chain, except for the well-known transverse coupling plasmons, propagating coupled quadrupolar plasmon mode which cannot be excited by light is also efficiently excited by electron beams. This plasmon mode exhibits a longer propagation distance along the chain and better spatial confinement. Meanwhile in the four-wire waveguide, the electron excited plasmons with zero momentum for each wire in the cross-section have longer propagation distance.

2. Calculation method

The interaction between metallic nanostructures and electron beams can produce energy loss which mostly comes from the excitation of plasmons, therefore electron energy loss can be utilized to explore surface plasmons. Specifically, the formula of EELS is defined as [14]:

$$\Gamma(\omega) = \frac{e}{\pi\hbar\omega} \int dt \, \mathrm{Re}\left\{ \mathrm{e}^{-\mathrm{i}\omega t} \mathbf{v} \cdot \mathbf{E}^{\mathrm{ind}}(\mathbf{r}_e(t), \omega) \right\}, \tag{1}$$

when an electron passing near a sample with constant velocity $\mathbf{v}$ along a straight line trajectory $\mathbf{r} = \mathbf{r}_e(t)$, and $\mathbf{E}^{\mathrm{ind}}$ is the induced electric field.

For the case of the coupled particle chain, the basic ingredients of particle chains under consideration are gold nanoparticles and their dielectric data is taken from Palik [15]. Collective plasmonic response of particle cluster arising from the interaction between neighbor particles can be investigated using either light or fast electrons as the excitation source. To obtain the quantity $\Gamma(\omega)$, the induced electric field is directly calculated using an exact electrodynamic multiple elastic scattering multipole expansions method (MESME) [16,17]. In contrast to other methods, such as dipole models or finite difference time domain simulations (FDTD), MESME is quasi-analytic method that includes all multipole expansion terms and converges fast to explore clusters. In brief, MESME expresses the fields scattered by each plasmon particle as a multipole expansion. The multipole excitations on each particle are driven both by the incident field generated by electrons and by the fields associated with all other particles, giving rise to a set of self-consistent equations for the multipole expansion coefficients that is solved recursively. Convergence results are achieved after inclusion of multipoles of orders $l \leq 11$.

And for the case of multi-wire waveguides, it is useful to decompose EELS into each parallel momentum $k_\parallel$ along the wire due to the translational invariance. Then the EELS probability is given by

$$\Gamma(\omega) = \int_0^\infty dk_\parallel \, \Gamma(k_\parallel, \omega). \tag{2}$$

with

$$\Gamma(k_\parallel, \omega) = \frac{e}{\pi^2\hbar\omega} \int dt \mathrm{Re}\{\mathrm{e}^{-\mathrm{i}\omega t} \mathbf{v} \cdot \mathbf{E}^{\mathrm{ind}}(k_\parallel, b, vt, \omega)\}$$

where

$$\mathbf{E}^{\mathrm{ind}}(k_\parallel, b, vt, \omega) = \int dx \mathrm{e}^{-\mathrm{i}k_\parallel x} \mathbf{E}^{\mathrm{ind}}(x, b, vt, \omega)$$

is the Fourier transform of the induced field along x, the electron trajectory is described by $z = vt$. We take z perpendicular to the wires, and b is the impact parameter of the electron beam relative to the wire axis. The quantity $\Gamma(\omega)$ can be directly calculated from the solution of Maxwell's equations using the boundary element method (BEM) [18], in which the electromagnetic field is expressed in terms of charges and currents distributed on the surface of the particles. Incidently, the propagation distance of SPPs usually defined as $l = [1/2\,\mathrm{Im}(k_\parallel)]^{-1}$, which represents the attenuation along the metal surface.

3. Propagating coupled-quadrupolar plasmon in a coupled nanoparitcle chain

Our coupled nanoparticle chain is shown in Fig. 1. It consists of two gold particle chains with diameter 160 nm and horizontal gap distance 5 nm, and the number of paticles in each chain is 51. The vertical gap distance between the chains is defined as d. An electron beam of energy 50

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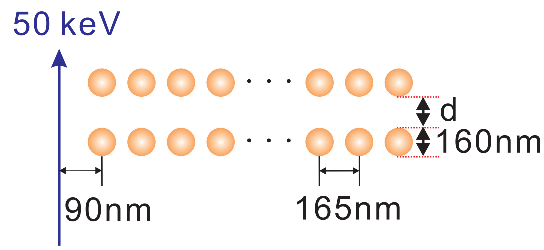

Fig. 1. Sketch of a coupled gold nanoparticle chain with an electron beam of 50 keV passing close to one of the end. The number of particle with diameter 160 nm in a single chain is chosen as 51, and the gap distance between horizontal neighbor particles is 5 nm.

keV travels parallel to one end of the particle chains at a distance of 90 nm. The excited SPPs propagate along the chain through the electromagnetic coupling between neighbor nanoparticles.

It is known that a nanoparticle chain supports longitudinal and transverse modes of propagating polarization waves [19]. However, only transverse plasmon mode is efficiently excited by electron beams in a single chain by fast electrons [10]. Now, we consider two nanoparticle chains, the interaction between neighbor particles extends to two-dimension from one-dimension. The photon energy resolved EELS is demonstrated in Fig. 2(a). It clearly shows that there are two maximum energy loss bands around 1.49 eV and 2.20 eV, which correspond to two types of plasmon excitation, respectively. The excitation probability for the low energy mode is increasing as the distance of gap d gets smaller, and almost vanishing as long as d is larger than 180 nm. Meanwhile, the higher energy band always maintains similar loss probability and does not show obvious dependence on the vertical gap distance. Therefore we can speculate that the bands with energy around 2.2 eV and 1.49 eV are related to the horizontal and vertical coupling between nanoparticles, respectively. The EELS spectrum at a vertical gap distance $d = 20$ nm is demonstrated in Fig. 2(b). And the induced electric near-fields and charge ordering of the two plasmon bands named as I and II are calculated and shown in Fig. 2(c) to further understand the nature of the excited plasmons. We can see that for the high energy mode II, the maximum values of the electric field only lie in the horizontal gaps between nanoparticles, which means that this mode corresponds to the transverse mode of propagating polarization wave. Whereas the electric field in the vertical gaps and horizontal gaps is always synchronous maximum for the low energy mode I, which indicates that it is a hybrid one with longitudinal coupling and transverse coupling between neighbor particles. Taking further analysising the induced charge ordering of the particle chains, surprisingly, we find that each metal particle is quadrupolar polarized for the low energy mode. One the other hand, we can also find that the decay of the high energy plasmon mode along the chain is much faster than the low energy mode. This can be explained by that each nanoparticle behaves like a dipole with high radiation losses in the high energy mode while like a quadrupole with low radiation losses in the low energy mode.

Next, we focus on the dispersion and dynamics of the excited plasmons in the coupled nanoparticle chains. The energy-resolved excited electric near-field in the vertical gap along the particle chain is sketched in Fig. 3(a). The low energy mode near 1.49 eV travels along the chain until the endpoint, while the high energy mode around 2.0 eV propagates along the chain but dissipates rapidly and almost disappears at 6 μm. This result is consistent with our previous analysis that the radiation losses are less in the low energy mode. Using Fourier transform to resolve the induced electric near-field in momentum space, we show the resulting dispersion curve of the excited plasmons in Fig. 3(b). The dispersion line lied on the right side of the light

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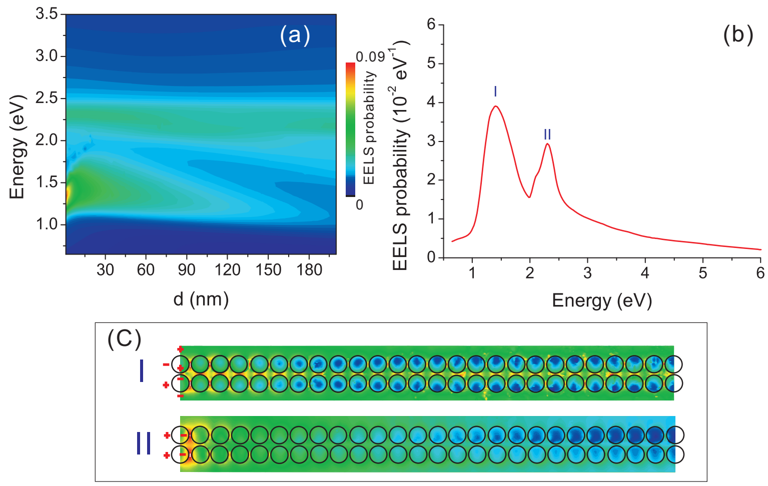

Fig. 2. (**a**) Photon energy resolved EELS probability dependent on the gap distance between two particle chains. Higher and lower energy bands shown by the maximum of energy loss are corresponding to horizontal coupled dipolar and quadrupolar plasmons, respectively. (**b**) EELS probability in the geometry shown in Fig. 1 with $d = 20$ nm. There exits two plasmon modes named after I and II with energy 1.49 eV and 2.2 eV, respectively. (**c**) The electric near-field distribution and the charge build-ups for plasmon modes I and II, respectively. Each particle shows quadrupolar distributions for mode I, whereas dipolar distributions for mode II.

line in vacuum demonstrates that the confinement of electric field near the surfaces of particles. Incidently, only one dispersion line is clearly shown because that the low energy mode is much more stronger than the high energy mode in this excitation geometry. To see time evolution of the excited plasmons propagating along the chain, the electric near-field in real space is obtained using time-domain Fourier transformation [Fig. 3(c)]. The dashed and dash-doted lines are used for guiding eyes, one can see that two plasmons are launched by the electron beam with different group velocities. The mode with slower group velocity but faster dissipation is the transverse coupling mode, while the other one which travels much faster and with longer propagation distance is the coupled quadrupolar mode. And there is a time difference between two plasmon modes due to finite velocity of electron beams. For our case, the speed of electrons is about 0.17c, and it takes about 5 fs for an electron to travel from $t = 0$ where electrons is at the center of the gap to the center of one single nanoparticle chain.

4. Higher-order propagating plasmons in a four-wire waveguide

In contrast with light coupling method, electron beams can also excite propagating plasmons in a single nanowire [10] and a coupled nanowire array [11]. In addition, swift electrons offer the benefit of exciting dark modes in nanowires compare to free light [20]. Accordingly, a symmetrical plasmon excitation scheme is designed [Fig. 4(a)]. We consider a symmetrical geometry formed by four silver nanowires and an electron beam of 100 keV passing through the center point of the structure, and the diameter of each wire is 160 nm with gap distance 10 nm. The dielectric data of silver wires is taken from Johnson and Christy [21]. The calculated wave-vector and photon-energy resolved EELS is shown in Fig. 4(b). The dispersion curves from the contour plot of EELS indicate that at least six kinds of plasmon modes are excited by the electron beam in this geometry.

The induced electric near-field for each mode is calculated and the corresponding induced

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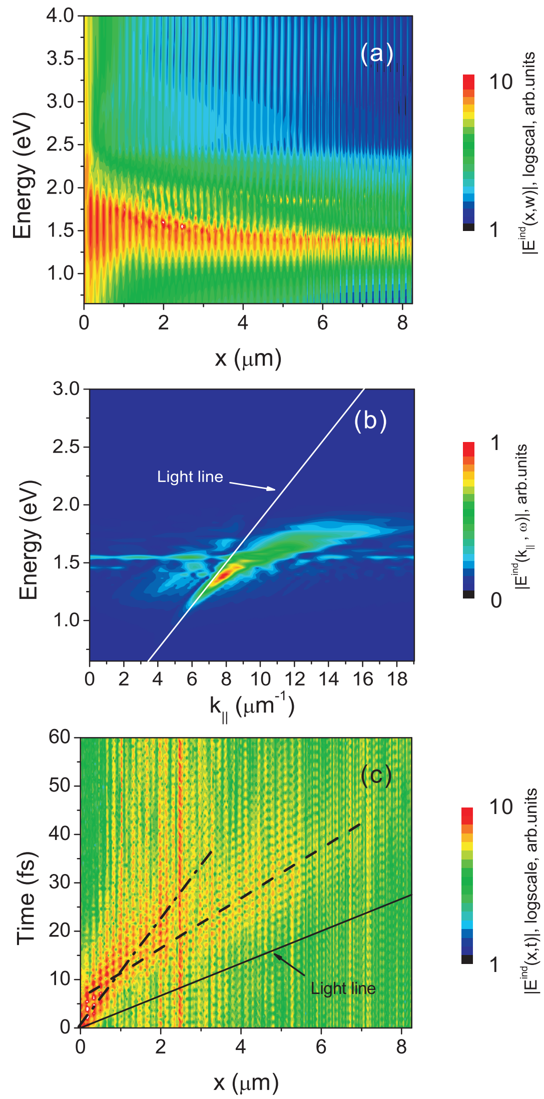

Fig. 3. Guided plasmons generation by an electron beam in a coupled nanoparticle chain. (**a**) Photon energy resolved electric near-field distribution along the chain in the transversal gap. (**b**) The dispersion curve of surface plasmons propagating along the chain. (**c**) Time evolution of excited surface plasmons.

charge ordering are analyzed [Fig. 4(c)] to unveil the nature of the excited plasmon modes. Taking use of the plasmon hybrid model [22], the excited plasmons can be divided into three classes: the mode labeled as A originates from the coupling of two gap plasmons, modes B-D are coupled plasmons with dipolar coupled plasmons in wire pairs, and modes E and F result from the hybrid of quadrupolar coupled plasmons. What we want to stress here is that the total dipolar momentums for modes B, E, and F are almost zero due to symmetry of the induced charge, which means that these plasmon modes can resist radiation losses compared to modes A, C and D.

To confirm this point, the propagation distances for each plasmon mode are calculated and

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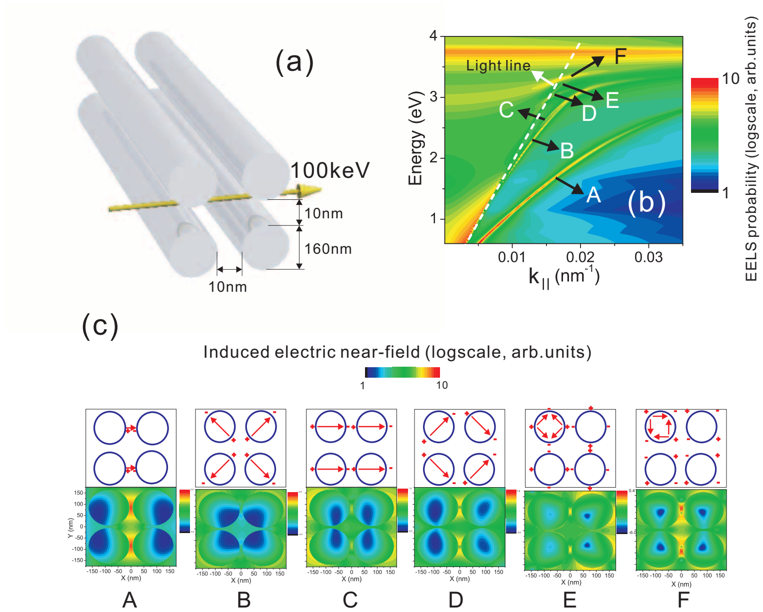

Fig. 4. Higher-order plasmons generation by electron beams in a symmetrical four-wire system. (**a**) Schematic of an electron beam of 100 keV passing thought the center point of symmetrical distribution of four nanowires with 160 nm in diameter, and the gap distance of neighbor wire is 10 nm. (**b**) Parallel momentum $k_{||}$ and photon energy resolved EELS probability. Propagating plasmons excited by the electron beam is label as A-F, respectively. (**c**) The induced electric near-field and charge ordering of excited plasmons in the cross-section of the wires.

compared in Table 1. We consider two different photon energies (1.55 eV and 3.3 eV) due to that excited plasmon modes A-F have different cut-off energies. At an energy of 1.55 eV, the plasmon mode B has the longest propagating distance up to 72.731 μm which is about seven times than the mode A and the mode C. This is consistent with that the total induced dipolar momentum of four nanowires is zero for the mode B, while the mode A and C possess non-zero total dipolar momentum. In addition, at an energy of 3.3 eV, the plasmon mode E holds the record for the longest propagation distance 1.322 μm which is almost ten times longer than mode A, and this in turn can be explained by the fact that each wire is almost "dipole emission forbidden". Although the propagation distance of plasmon mode F is less than mode E, it is also much longer than other bright modes (A, C and D). Therefore we can conclude that it is a practical way to use electron beams to generate low radiation losses propagating plasmon modes in a multi-wire system, eg. modes E and F in the four-wire system.

Table 1. Comparison of Propagation Distances for Excited Plasmons by Electron Beams in the Excitation Scheme Fig. 4(a)

Plasmon mode label	A	B	C	D	E	F
Propagation distance (μm)|1.55 eV	11.092	72.731	9.670	×	×	×
Propagation distance (μm)|3.3 eV	0.1298	0.760	0.128	0.212	1.322	0.686

5. Discussion and conclusion

Traditionally, although with very high confinement of electromagnetic field, a nanoparticle chain is though as a non-efficient plasmon guiding structure due to the scattering light from surface plasmon, and the propagation distance of SPPs is always below 1 μm. And determining how to suppress the radiation losses is important for practical applications. In our case, fast electrons offer one possible solution for this problem, the coupled quadrupolar modes with approximately zero total dipolar momentum in each nanoparticle can efficiently reduce the radiation losses. And this cannot be realized by free radiation due to the momentum mismatch between them. At the same time, the velocity of electrons can be easily controlled, thus provides us another dimension to control surface plasmons, which may be used as an ultrafast detection means. Not only in nanoparticle chains, electron beams can also excite higher, multipolar plasmon in multi-wire waveguides based on the same principle, therefore one can use fast electron beams as a long propagation plasmon source. And these kinds of plasmons, being insensitive to excitation by light, can encode information that cannot be extracted out in the form of light and may offer an advantage in some circumstances.

To conclude, in this paper we proposed to use electron beams to excite higher-order, multipolar plasmons to reduce radiation losses in coupled nanoparticle chains and multi-nanowire waveguides. In the coupled nanoparticle chain, the guided plasmon mode with quadrupolar particles can be excited by fast electrons, then achieves long propagation distance. Meanwhile, in the multi-wire waveguide, propagating plasmons excited by electrons with zero-momentum in the transverse cross-section possess longer propagation distance due to inhibition of radiation into far-field. And these properties make fast electrons possible more useful in communication technology.

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