Plasmon-suppressed vertically-standing nanometal structures

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Abstract: The authors report plasmon-suppressed vertically-standing nanometal-stripe-array structures fabricated by Ar ion sputtering after electron-beam lithography and Ag deposition. When the width of the Ag stripe is comparable to the skin depth of a metal (~ 20 nm), the particle plasmon resonance is strongly suppressed for electric fields oscillating perpendicular to the length of the stripe. This suppression of the particle plasmon excitation is attributed to the limited movement of free electrons localized near the bottom of Ag stripe. This plasmon-suppressed vertically-standing nanometal structures could be used for broad band polarizers.

OCIS codes: (250.5403) Plasmonics; (310.6628) Subwavelength structure, nanostructures

References and links

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1. Introduction

As nanotechnologies have matured rapidly in recent years, various methods have been tried to fabricate nano-scale metallic structures. The most common method employs dry etching techniques after electron-beam lithography [1]. The lift-off technique is another famous method to define patterns utilizing the electron-beam lithography and metal deposition [2].

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The focused ion beam etching method is also used to fabricate metallic nano patterns directly [3]. In addition, self-assembled metallic rings based on the sputter re-deposition were reported recently [4]. However, in the case of the self-assembly, it is nontrivial to generate desired nano-patterns at specific positions. In this report, novel vertical nanometal structures prepared by Ar ion milling and electron-beam lithography are investigated.

The plasmonic resonance appearing in metallic structures [5] has attracted many researchers because of a wide variety of applications such as surface plasmon resonance sensors [6], surface-enhanced Raman spectroscopy [7], and second-harmonic generation [8]. However, the spectral dependence of the plasmonic resonance prevents these metallic structures from their application to broadband polarizers [9]. Here, we propose and demonstrate vertically-standing nanometal-stripe-array structures in which the plasmonic resonance is strongly suppressed over the entire visible spectrum. The optical characteristics are studied by polarization-dependent transmission measurement. The interaction between the free electrons and the localized plasmonic field is analyzed by the finite difference time domain method to understand the physical origin of the suppression of the plasmon resonance.

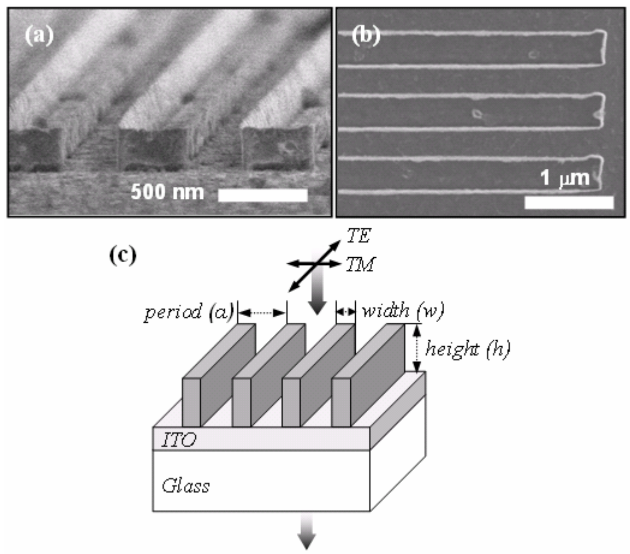

Fig. 1. SEM images of fabricated vertically-standing gold nanometal structures: (a) side view (60°) . (b) top view. (c) schematic of nanometal stripe array.

2. Sample fabrication and experimental setup

The vertically-standing nanometal-stripe-array structures are prepared as follows. First, a 125-nm-thick indium tin oxide (ITO) layer is prepared on glass substrate to avoid charging effects during the electron-beam writing. Next, Polymethyl Methacrylate (PMMA) is spin-coated and one-dimensional grating structures are defined on the PMMA layer by the electron-beam lithography. And then, Ag (or other metals) is deposited over the grating structure by thermal evaporation. During the subsequent Ar ion milling process, the deposited Ag is removed and simultaneously sputter-deposited on the side walls of the PMMA. Finally, the sample is covered with PMMA overcoat to prevent the Ag film from oxidation.

The fabricated structure just before the protective PMMA overcoat is shown in Fig. 1(a). Typically, the width of the stripe is ~ 20 nm, which can be controlled by the thickness of the initial metal layer. The height of the stripe is 200 nm, the same as the thickness of the PMMA layer. The total size of the pattern is 50 μm × 50 μm. A white light (halogen lamp) transmission setup is used for experiment. A Glan-Thomson polarizer is placed in front of a sample to control the polarization state of the input light. Two representative directions of electric fields, oscillating parallel (TE-like polarization) and perpendicular (TM-like

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polarization) to the stripe axis, are chosen for our measurement and defined as described in Fig. 1(c). Because of the small size of the pattern, the transmitted beam is magnified and the light passing through the patterned area is sampled by a pin hole. The spatially-filtered light is collected into the entrance slit of the Optical Multi-channel Analyzer. The transmission spectrum taken from the same wafer without the pattern is used as a reference.

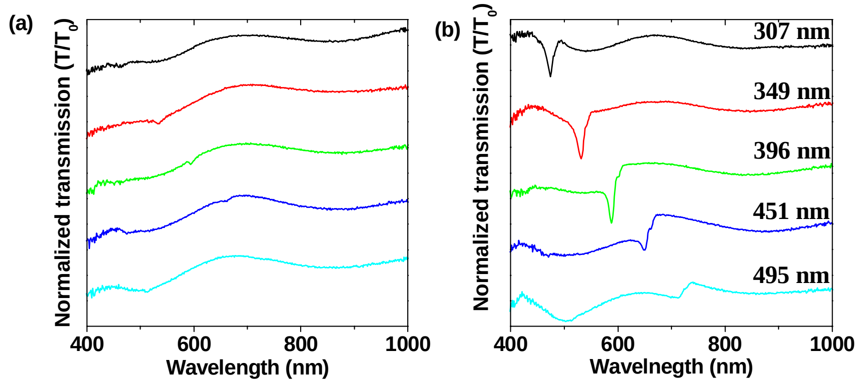

Fig. 2. Normalized transmission spectra of vertically-standing Ag nanometal stripe arrays of different periods: (a) TM-like polarization and (b) TE-like polarization. The width and the height of Ag stripes are 20 nm and 200 nm, respectively. The number in (b) is the period of the array.

3. Results and discussion

In periodic metallic structures, the particle plasmon resonance is easily observed with the help of the Bragg momentum [9, 10]. In order to study the particle plasmon resonance in our vertically-standing Ag nanometal stripe arrays, polarization-dependent transmission spectra are taken at normal incidence as a function of period. Figure 2(a) shows normalized transmission spectra of TM-polarized light. Unlike in the conventional metal stripe array, no noticeable plasmonic resonance is identified over the whole visible spectrum. In addition, the transmission spectra show no dependence on the period of the array. In other words, the vertically-standing Ag nanometal stripe arrays do not seem to interact with TM-like fields. However, in the transmission spectra of TE-like fields, unambiguous resonant lines are easily observed as shown in Fig. 2(b). These asymmetric resonances are identified as the Fano-resonance, a kind of Wood's anomaly related to the ITO guided mode [10]. In this case, the wavelength of the Fano-resonance varies with the grating period.

The electric field of a particle plasmon resonance is highly concentrated at the metal/dielectric interface. Therefore, the width of Ag stripe plays an important role for the plasmonic excitation [9]. To investigate the width dependence, we prepare Ag stripes of various widths as shown in Fig. 3(b). Here the nanometallic pattern repeats at a fixed interval of 340 nm. The vertically-standing Ag stripe (w = 25 nm) is 125-nm high and over-covered with PMMA. Those comparison samples fabricated by conventional lift-off techniques are smaller in height (55-nm). In the case of TM-like polarization (Fig. 3(a)), the transmission spectra show noticeable dependency on the width of the Ag stripe. When the width is 141 nm, the particle plasmon resonance near 600 nm is broad and strong. As the width decreases, the resonance becomes narrower and weaker. It is interesting to observe that the plasmon resonance almost disappears in the vertically-standing Ag stripe array. Also note that the particle plasmon resonance red-shifts with the increase of the width of the metal. In comparison, in the case of TE-like light (Fig. 3(b)), the spectral position of the Fano-resonance is fixed near 540 nm independent on the width of the metal. It is not unexpected because the resonance of the ITO-guided mode depends only on the period of the metallic grating. When the width of the Ag stripe decreases, the Fano-resonance becomes weaker because the scattering cross section is reduced. However, the asymmetric line shape is still maintained even in the very thin vertically-standing structure.

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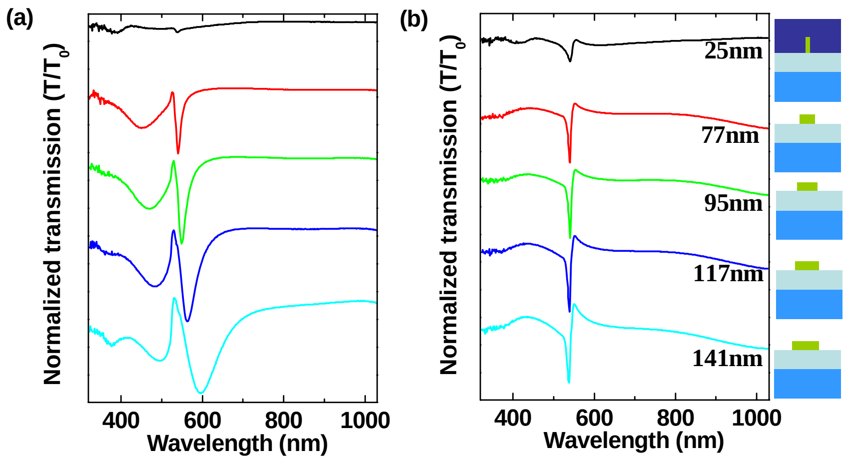

Fig. 3. Normalized transmission spectra of various widths of Ag stripe arrays: (a) TM-like polarization, (b) TE-like polarization. The numbers in (b) are the widths of the Ag stripe. The pictures on the right of (b) are the vertical cross sections. The Ag stripe of w = 25 nm has 125-nm height with PMMA over-coat. The other samples are 55-nm high without PMMA over-coat.

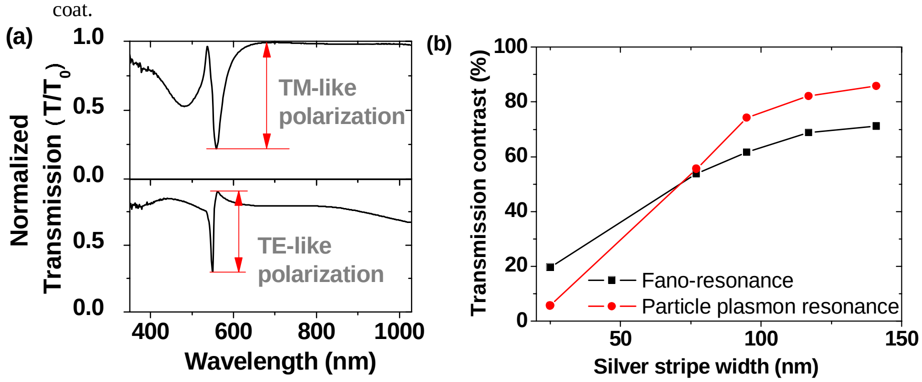

Fig. 4. (a) Description of the transmission contrast of the resonance. (b) Transmission contrasts of Ag stripe arrays as a function of the width. The particle plasmon resonance and the Fano-resonance are considered for TM-like and TE-like polarizations, respectively.

To study the width dependence, transmission spectra near resonances are plotted as shown in Fig. 4. The maximum reduction of the transmission is one way to represent the strength of the coupling between the incident photons and the resonant mode. In this study, we define the transmission contrast as the ratio of the minimum transmission and the maximum transmission as indicated in Fig. 4(a). It seems natural that the transmission contrast increases with the increase of the width of the metal. However, it is worth pointing out that the transmission contrast of the particle plasmon resonance diminishes more rapidly than that of the Fano-resonance. The periodic Finite-Difference Time-Domain (FDTD) method [11] is employed to understand our observation. In these analyses, the stripe width is varied from 10 nm to 141 nm, while the height and period of the Ag stripe are fixed at 55 nm and 340 nm, respectively. As shown in Fig. 5, the computed transmission spectra (solid lines) compares reasonably with those of the measurement (dotted lines). In the case of the Fano-resonance, the transmission contrast shows a rather large value even when the width is 20 nm and becomes saturated when the silver stripe width is over 40 nm. In comparison, the transmission contrast of the particle plasmon resonance drops rapidly when the width becomes smaller than 60 nm and becomes weaker when the width is 20 nm. It is worth remembering that the particle plasmon resonance disappears when the dimension of the metal width is comparable to the skin depth (25 nm) of Ag metal [12].

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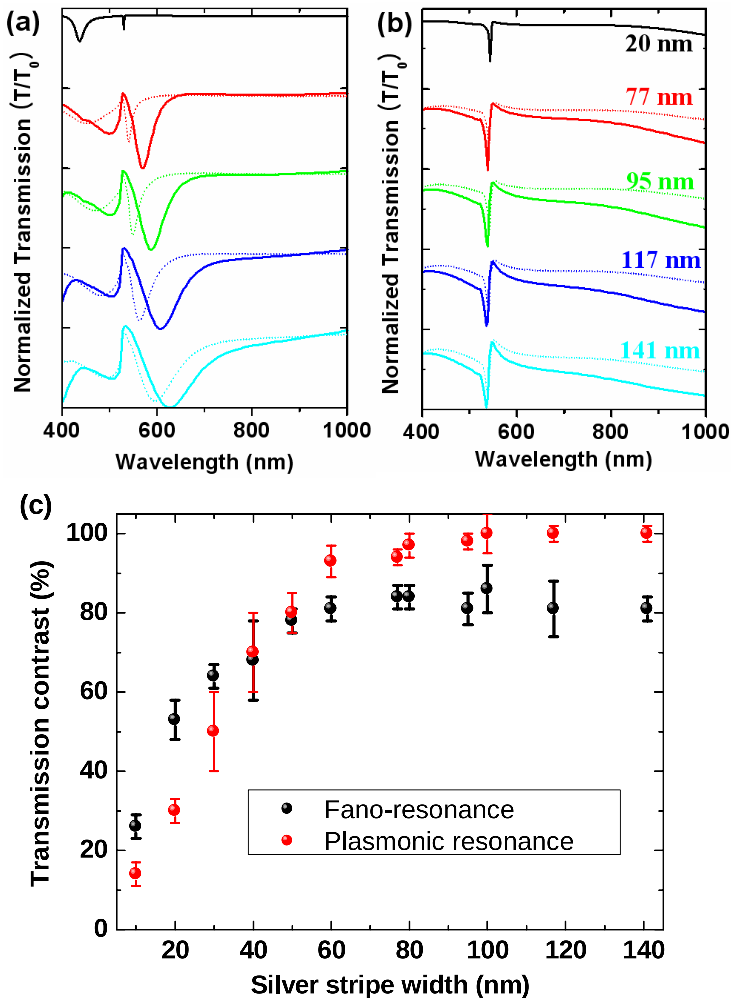

Fig. 5. Calculated normalized transmission spectra and transmission contrasts for Ag stripe arrays of various widths. (a) TM-like polarization, (b) TE-like polarization, (c) Transmission contrasts as a function of width. The dotted lines are the experimental data. The numbers in (b) indicate the width of Ag stripes.

The electromagnetic field distribution and the induced current density are analyzed by FDTD method. The induced current density, *J(t)*, is calculated using the equation shown below [11],

$$\frac{\partial \vec{J}(t)}{\partial t} + \gamma \vec{J}(t) = \varepsilon_0 \omega_p^2 \vec{E}(t)\,. \qquad (1)$$

Here, the plasma frequency, ω_p is $1.155 \times 10^{16}\ s^{-1}$ and the electron-neutral collision frequency, γ is $1.108 \times 10^{14}\ s^{-1}$. These values are estimated using the data in the reference [13] and the resultant Drude model generates optical constants that deviate less than 5% from the reported data over a spectral range of 400 nm ~ 800 nm. Figure 6(a) shows the calculated transmission spectra of the vertically-standing Ag stripes with thickness of the skin depth of 25 nm formed on 125-nm-thick ITO film. The particle plasmon resonance appearing at 488 nm is much weaker than the Fano-resonance at 558 nm. For TM-like light, the dipolar E_z field is strongly localized near the bottom edge of Ag stripe as shown in Fig. 6(b). The induced current is highly concentrated at the bottom of the Ag stripe, where the electrons oscillate perpendicular

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to the stripe (x-direction). In this case, the movement of the electron is severely restricted by the finite metal width on the order of the skin depth. In other words, the electron motion perpendicular to the surface 1D structure is more restricted in comparison to that along the surface of the 3D structure. Estimating from the data shown in Fig. 4, the width of Ag stripe needs to be at least more than twice the skin depth for electrons to move freely. The transmission of TE-like light decreases for longer wavelengths as shown in Fig. 6(a). However, note that the transmission of TM-like light maintains a value close to one. As the width of the metal increases with a fixed period, the extinction ratio between two polarizations gets better as one can see in Fig. 7.

On the other hand, the Fano-resonance still remains relatively strong. As shown in Fig. 6(c), the H_z field component of this Fano-resonance is mostly confined within the ITO layer. This is one of the evidences that the Fano-resonance is Wood's anomaly related to the ITO guided mode. Moreover, electrons oscillate freely along the length (y-direction) of the stripe and the induced current spreads out without restriction. Note that the electromagnetic field becomes weak inside the metal film. Unlike at the particle plasmon resonance, the metal width comparable to one skin depth provides enough room for electrons oscillating parallel to the stripe.

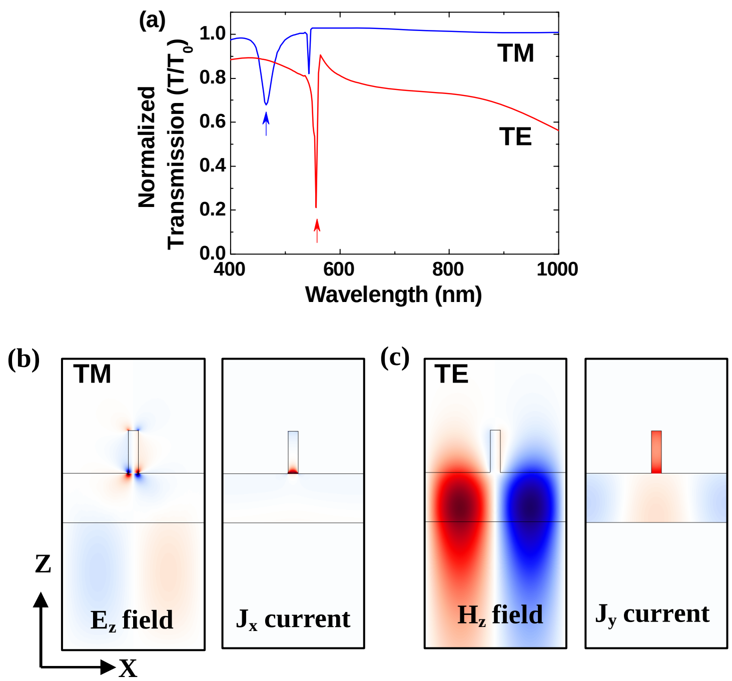

Fig. 6. (a) Normalized transmission spectra of vertically-standing Ag stripe array at TE-like and TM- like polarizations. (b) E_z field distribution and J_x current distribution for the particle plasmon resonance at 488 nm (the blue arrow in (a)). (c) H_z field distribution and J_y current distribution for the Fano-resonance at 558 nm (the red arrow in (a)).

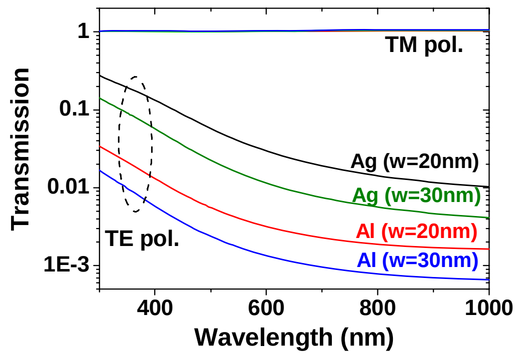

Fig. 7. Transmission spectra of vertically-standing metallic stripes. The period is 200 nm and the height of the stripe is 200nm. The black and the green lines indicate Ag stripes of 20-nm width and 30-nm width, respectively. The red and the blue lines indicate Al stripes of 20-nm width and 30-nm width, respectively.

4. Summary

In summary, we fabricate plasmon-suppressed vertically-standing nanometal structures by the electron-beam lithography and Ar ion sputter redeposition. In this novel structure, the particle plasmon excitation is strongly suppressed for TM-like light. However, the Fano-resonance is still observed for TE-like light. We conclude that the localized interaction between the electromagnetic field and the free electron oscillation is responsible for the disappearance of the particle plasmon when the width become comparable or smaller than the skin depth. The plasmon-suppressed vertically-standing nanometal structure could be a good candidate for a broad polarizer over the entire visible spectral range.

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