Engineering localized hotspot both in transversal and longitudinal direction by plasmonic coupling between nano-particles and reflective metallic film

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A structure which consists of photoresist film sandwiched by Ag nano-particle and metal film is proposed to modify localized hotspot both in transversal and longitudinal direction. It shows that there is strong plasmonic coupling between Ag nano-particle and metallic surface, which helps to reduce the width and elongate the depth of the plasmonic hotspot localized inside photoresist film. And that fringes and side lobes around hotspots can be effective attenuated by bottom-side illumination. Influences of illumination, particles inter-space, and polarization are also studied. The method opens avenue for the potential applications such as lithography, optical storage, etc.

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One of the most remarkable phenomenon associate with metallic nano-particles at optical wavelength is field enhancement effect[1]. And the induced localized optical field within metal nanostructures usually displays strengths orders of magnitude than the incident light and strong confinement in nano dimensions[2]. This occurs mainly due to the excitation of surface plasmon modes at visible frequencies. A number of applications have been explored by engineering such properties like surface enhanced Ramam scattering[3], nano lithography[4,5], bio-sensor nanolithography[6,7], and optical storage etc. Among these investigations, great deals of efforts have been focused on the problems of increasing the enhancement factor or reducing the spot size for improved sensitivity or spatial resolution[8,9]. For some applications, however, the hotspot's light distribution features in three dimensional spaces (like focus depth, side lobe lever etc) usually plays a dominating role in determining the property of recorded nano patterns. This issue becomes serious especially for nano-particle lithography, in which hotspots localized in the vicinity of the nano-particles are damped quickly as going away, resulting in considerably decreased resolution and shallow feature profiles of the photo-resist.

Here we introduce a simple method to modify localized hotspot both in transversal and longitudinal direction as a result of coupling between nano-particles and metal film. The structure consists of photoresist film sandwiched bynano-particles array and metal film. Unlike traditional particle lithography, which light illuminates sample uprightly, we demonstrate a new method with illumination direction inverted. Our numerical simulation results show that the fringes around the spot can be effectively eliminated by this inverted illumination method, and the spot size can be compressed transversally and elongated longitudinally by the effect of plasmonic coupling, which is because of compensation effect along the longitude direction.

The schematic of the structure is shown in Fig. 1(a). It consists of a group of Ag nano-particles positioned above photoresist film as an insulator layer. Beneath the photoresist is Ag metal layer deposited on the SiO_2 substrate. The distance between the particles and the surface of photoresist is assumed to be zero here. Monochromatic plane wave light in the normal direction and circular polarization is incident beneath the substrate.

The thickness of Ag film is assumed to be small enough for light transmission through the penetration effect. So beneath every nano-particle, a hot spot of light could be observed, and this is usually attributed to the "lightening rod effect"[10,11], which concentrates a great number of charges around the particles apex region and local field enhancement. In order to get insight into the physics of particle-sample resonant coupling, a simple quasi-electrostatic model can be effectively used. As shown in Fig.1(b), the Ag spherical particle located at the photoresist surface, having refractive index $\varepsilon_{\rm m}$, radius $a \ll \lambda$. When it is illuminated by the incident field E_0, a dipole $\mathbf{p} = \alpha_{\rm p}\mathbf{E}_0$ is generated, in which $\alpha_{\rm p} = a^3\frac{\varepsilon_{\rm m}-1}{\varepsilon_{\rm m}+2}$[12] is the polarizability of the particle. On the other hand, the Ag film with photoresist layer provides the possible existence of surface plasmon resonance

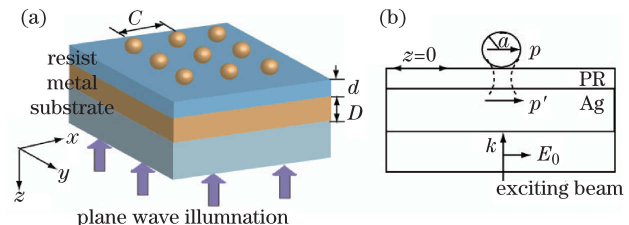

Fig. 1. (Color online) (a) Schematic of the nano-particle lithography structure, the curve radius of the sphere is 25 nm, and the particle inter-space C is 200 nm, the photoresist and Ag thickness are d=35 nm and D=40 nm, respectively; (b) Sketch of the particle-sample interaction model.

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as $Re[\varepsilon_\text{m}] \approx -\varepsilon_\text{pr}$, which occurs for light wavelength around 365 nm. So the excited dipole of particle would generate the image dipole inside the metal film. To keep the nano hotspot feature, the photoresist thickness should be small and evanescent waves of dipole excite surface plasmon, which form the strong coupling mode and engineer the hotspot shape both in transversal and longitudinal directions. In addition, Ag film acts as an optic attenuator for the bottom incoming light.Although it would decrease the hotspot peak intensity, the reduce

Fig. 2. (Color Online) (a) and (b) are spot size (FWHM) and electric filed enhancement ratio as functions of different position in photoresist when there is/isn't Ag film. (c)–(e) show electric charges density distribution at the top and bottom surface of the photoresist film when the incident light is (c), (d) circular polarization and (e) linear polarization, and the Ag film is removed in (c).

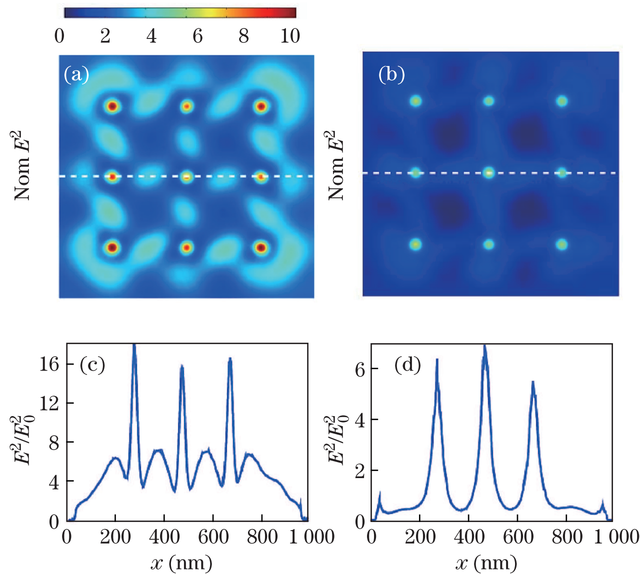

Fig. 3. (Color online) Electric field distribution at 15 nm below the top surface of photoresist film with 3×3 particles array structure with the normal plane light illuminated from (a) the particle side and (b) substrate side. (c) and (d) show cross curve of dot-line in (a) and (b) respectively, the reference plane is in the middle of the photoresist film.

of fringes or side lobes around hotspot makes the optic field much more distinguishable.

To give deep understanding of the hotspot modification due to the coupling of the particle-dipole and plasmonic reflector, numerically simulation in three-dimensional (3D) model is performed by COMSOL Multiphysics 4.2a. The Ag nano-particle radius is assumed to be 25 nm and the thickness of photoresist and the Ag film are d=35 nm and D=40 nm, respectively. For incident light at wavelength of 365 nm, the relative permittivity of Ag, photoresist and SiO_2 substrate are $\varepsilon_\text{Ag} = -2.4012 + 0.2448*i$[13], $\varepsilon_\text{pr} = 2.56$, and $\varepsilon_{\text{sio}_2} = 2.13$[14], respectively.

Figures 2(a) and (b) show the full-width at half-maximum (FWHM) and electric field enhancement ratio (E^2/E_0^2) variation along the depth direction in the photoresist. And in order to demonstrate the role of the Ag film, the contrastive result without the Ag film (black line) is presented as well. As shown in Fig. 2(a) and (b), the spot size enlarges greatly from ∼28 nm just below the particle to ∼110 nm at photoresist bottom surface, and at the same time light intensity (I=$|E|^2$) drops to about 1/10 times of the value of the interface when there is not metal film beneath the photoresist. However, the spot size is controlled within ∼60 nm (λ/6) and the attenuated intensity gets compensation when the metal film is added (red line). It is worth to note that the spot width inside photoresist is mainly determined by the resist thickness. So we can obtain spot size even below 20 nm, provided that the metal film and resist thickness are optimized.

To demonstrate the coupling behavior and its role in engineering hotspot shape, the electric charges density distribution at the top and bottom surface of the photoresist film is presented in Fig. 2(c) and (d). It is can be found in Fig. 2(c) that a greatly enhanced electric charges density appears with dipole distribution in the top surface of the photoresist film. At the bottom surface, however, the excited image dipole electric chargesare almost imperceptible (It is emphasized in Fig. 2(c)). As shown in the Fig. 2(d), when there is 40-nm Ag film added below the photoresist film, the dipole electric charges density in the bottom surface is increased ∼20 times compared with that in Fig. 2(c). Additionally, the orientation of dipole pattern rotates slightly due to the circular polarization of incident light (Fig. 2(c) and (d)). This phenomenon does not occur when the illuminating light is linear polarization (Fig. 2(e)). This can be easily understood since the polarization state evolves in the light propagation from the top to the bottom surface of photoresist.

As is well known, a great number of nano-particles are usually employed in 2D/3D array to increase its lithography patterns size. Influences from the illumination direction, the spacing distancebetween particles, and the incoming light polarization are discussed below.

To demonstrate the influence of illumination direction, we consider a 3×3 nano-particles array. The space between the particles is fixed at 150 nm. In Fig . 3(a) and (c), the plane wave with circular polarization is incident from the nano-particles side. The light interference between the particles results in great fringes around hot spots with about $6E_0^2$ side lobe intensity (37% of the

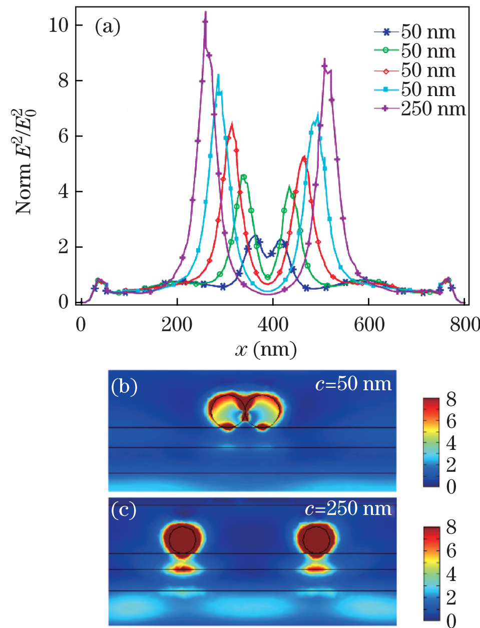

Fig. 4. (Color Online) (a) For two Ag particles, electric field enhancement ratio at 15 nm below the photoresist/particle interface when the particle inter-space c is 50, 100, 150, 200, 250 nm. (b) and (c) show the electric intensity when c is 50, and 250 nm.

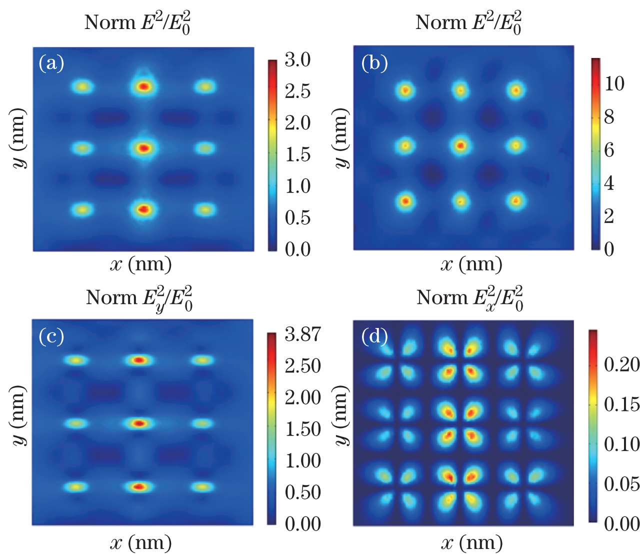

Fig. 5. (Color Online) (a) For a 3×3 nano-particles array (period is 150 nm), numerical simulation electric field enhancement ratio of linear polarization incident light with Hx=1, Hy=0. And its component in the (b) x direction and (c) y direction. (d) compared result of circular polarization incident light with Hx=1, Hy=i.

hotspot peak intensity $16E_0^2$). In Fig. 3(b), light is incident normally from the substrate side. In this case, the intensity of interference pattern is absorbed due to the obstacle of Ag film. The localized hotspots, however, are excited due to the coupling of light on the two sides of Ag film, which delivers about just weak fringes intensity (about 7% of hotspot peak).

Spaces between nano-particles are another important factor influencing the shape and intensity of hotspots. To see this clearly, only two adjacent particles with centers to center distance c are considered here. As shown in Fig. 4(a), the intensity enhancement ratio decreases as two particles get closer. When the two particles contact to each other (c is 50 nm), the intensity profile of two hot spots is almost undistinguishable. This occurs because the excited surface plasmon light is mainly localized between the space of the two particles (Fig. 4(b)), and the Ag film-particle coupling effect is correspondingly low in this case. As the two particles ares eparated, the hotspot between the two particles gradually weakens, instead, coupling and excitation of hotspots between the particle and Ag film becomes strong.

In Fig. 5, we present the calculation results with the linear and circular polarized illumination light. As shown in Fig. 5(a), the hot spot widens at the x axis direction due to the x polarization and the hotspots become ellipse. The electric field intensity ($|E|^2$) has 3 times enhancement in this case and Ey component is the dominant field (about 95% shown in Fig. 5(c)). The small quantity of excited Ex component forms the deleterious background noise (Fig. 5(d)). It can be found that the middle lines of the dots have greater enhancement than the both sides. This is because the middle particles carry an excitation more than the particles in the two endpoints[4]. But, this unequal excitation can be eliminated by illuminating with circularly polarized light. As shown in Fig. 5(b), the hot spot is a circular shape and Ex, Ey component reach 50%, respectively.

In conclusion, a new method for modifying hotspots associated with plasmonic effect between particles and metal films is introduced and demonstrated by numerical simulation. With inverted illumination, the localized electric field with dipole distribution mode is excited in the photoresist film, and forms a strong plasmonic coupling between the Ag nano-particle and metallic surface. Compared with metallic particles without reflective films, this new design possesses unique advantages of engineering hotspots both with reduced spot size and elongated depth inside the photoresist. In addition, the side lobe and fringes of light could be attenuated. This structure may play an important role for the future applications of nano lithography, optical storage, etc.

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