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Trapping nanospheres within graphene-based heterogeneous plasmonic nano-trench

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Abstract

We theoretically investigate a graphene-based heterogeneous plasmonic nano-trench for trapping noble metal and non-metal nanospheres. We propose heterogeneous plasmonic modeling geometry consisting of silver, gold and graphene for functioning plasmonic trapping operating at the near-infrared spectrum. For our designed model, the vertical potential well is 32 times larger than the Brownian motion energy K_BT, indicating that the nanosphere can be stably trapped using the graphene covered gold and silver nano-trench structure. By varying the incident laser wavelength and angle of illumination, the gold nanosphere can be trapped stably 40 nm above the bottom of the proposed nano-trench structure. The graphene layer enhances the localized electric field of the nano-trench in comparison to a pure silver and gold nano-trench structure without a graphene layer on top.

Keywords: stable plasmonic trapping, heterogeneous nano-structure and graphene layer

(Some figures may appear in color only in the online journal)

1. Introduction

Stable trapping of particles on nanoscale with optical tweezers can be highly expected for a wide range of applications in fields of light trapped assisted nanopatterning [1], DNA sorting [2], bio-sensor [3], and nanoimaging [4]. Efforts towards high-precision trapping of nano-scale targets have been developed recently, which is called plasmonic nano-trapping [5]. Generally, the plasmonic nano-trapping can be supported by plasmon resonance of several types of structures such as nano-dots, nano-pillars, dipole nano-antennas, nano-bowties, and nano-apertures [5–9]. Amongst them, the U-shaped trench plays a key role in supporting the nano-scale electromagnetic (EM) field for supporting the strong plasmon resonance [2], which exhibits huge potential in exciting the highly localized optical near field for nano-trapping realization. Unfortunately, it usually leads to the plasmon off-resonance of the U-shaped trench geometry due to the fabrication ability limits and laser irradiation instabilities [2]. As a result, the stability of plasmonic nano-trapping can be deteriorated, bringing large challenging towards the high-precision plasmonic trapping of nanoparticles. Recently, graphene sheets, one-atom-thick two-dimensional layers of sp 2-bonded carbons, were predicted to have a range of unusual properties [10]. Especially, the graphene plasmon can be expected to bear the tunability for manipulation of the nano-trapping [11]. As the graphene-based materials are applied, the plasmonic resonance spectrum for determining the trapping wavelength can be potentially modified for advancing a wide

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range of applications [12]. Although the graphene can bring in the potential benefit of tuning the plasmonic nano-trapping spectrum, the trapping stability can still be deteriorated in case of nanoscale particles due to the non-deterministic scattering cross-sections as considered in different shapes and sizes, made of different materials [10–12]. A systematical investigation of well-defined nano-trapping is highly expected for understanding of the specific trapping properties of the graphene heterogeneous nano-trench as considering typical trapping target nanoparticles made of a wide range of materials.

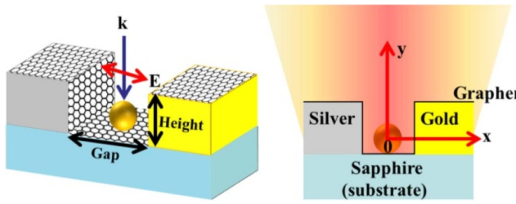

Figure 1. Schematic of the heterogeneous U-shaped trench with trapping gold nanosphere. The heterogeneous nano-trapping U-shaped trench is immersed in water (refractive index (n) of 1.33). The direction of the incident light is represented by wave vector k, which is vertical to the surface of silver and gold film. 'E' denotes the polarization of incident light. The U-trench gap/height changes from 200 nm to 600 nm.

2. Theoretical model

In this paper, the graphene-based heterogeneous U-shaped trench geometry for stable nano-trapping of different nanospheres is investigated numerically based on the finite element method (FEM). A 2D plasmonic model for predictions of plasmonic trapping is proposed, in which the designed nano-trapping geometry is made of heterogeneous noble metals of silver and gold films and covered graphene on top. We observed that a wide range of nanospheres can be flexibly manipulated in water environment and specific gold nanospheres can be stably trapped in the gap of the graphene covered nano-trench. The stable nano-trapping is carefully investigated with respect to the key role of graphene participating in enhancing the localized electric field compared to the pure silver and gold geometry. Moreover, the current design limit is the comparative tunability ability due to mono-layer graphene sheet. In fact, the tunability can be further improved by using few layer graphene (FLG) instead of mono-layer graphene, which is out of the scope of the current work. In this work, we mainly focus on the stable nano-trapping of 2D U-shaped trench using graphene of thickness 0.335 nm for well definiing the stable trapping of very small nanosphere (40 nm sphere) within the U-shaped trench covered by mono-layer graphene. Also, we proposed the silver/gold and graphene for plasmonic trapping operating at an even wider range of near-infrared spectrum compared to that without graphene. The strongly interacting plasmon resonance can exist in heterogeneous structures compared to the homogenous (silver/silver or gold/gold) ones, which have shown potential advantages over the homogeneous structures in many aspects such as broadening the resonance spectrum or leading to a stronger enhancement in the gap of U-shaped trench. Moreover, the model is fully consistent for field calculation, accounting for the presence of the nano-sphere or perturbative (calculated without the nano-sphere). In fact, the trapping sphere is put in the U-trench, and the electric field can be affected by the sphere itself. The current model can work for the whole space calculations of the e-field distributions based on the full-space Helmholtz EM equation. The schematic of the heterogeneous U-shaped trench geometry for trapping gold nanosphere is shown in figure 1. The design consists of a U-shaped trench made on a silver/gold metal heterogeneous film on a sapphire substrate with a graphene layer on top. Graphene with a specific thickness (a mono-layer) is coated on the top of U-trench structure. Physically, as a light-wave with wave vector 'k' is incident on the U-shaped trench as in figure 1, the electron systems of the silver, gold and graphene are synchronously excited initially in the heterogeneous system [13]. The heterogeneous system gets electronically resonant as laser wavelength matches the surface plasmon frequency determined by the specific trench geometry and the graphene electron energy level. The gradient distribution of localized electric field can be dominant in generating the trapping force within the U-trench gap. In this model, the optical forces applied on the nanosphere can be calculated as [14]:

$$\mathrm{F} = \frac{1}{4}\varepsilon_0 \mathrm{Re}\{\alpha\}\nabla|\mathrm{E}|^2 + \frac{n\sigma}{2\mathrm{c}}\{\mathrm{E}\times\mathrm{H}^*\} + \frac{\sigma}{2}\mathrm{Re}\left\{\mathrm{i}\frac{\varepsilon_0}{\mathrm{k}_0}(\mathrm{E}.\nabla)\mathrm{E}^*\right\} \tag{1}$$

where ε_0 is the permittivity of the free space, σ is the total cross-section of the nanosphere, c is the speed of light, E is the electric field, k_0 is the wave vector in the free space and H is the incident magnetic field. The first term is the intensity of gradient forces that facilitates spatial confinement in optical tweezing, which also dominates the other two forces. The second term is radiation pressure force and it corresponds to a force in the light propagation direction and 'n' is the refractive index of surrounding ambient. The third and final term gives the polarization force. Furthermore, the polarizability is a key factor that characterizes optical response due to the interaction between the optical field and the nano-structure that determines the strength of interaction. In this model, sphere radius 'α'' is defined for polarizability. Assuming a metallic spherical Rayleigh sphere with a radius of 'α' suspended in a medium with the dielectric constant of 'ε', its optical property can be described by the polarizability 'α'' [14]:

$$\alpha = 4\pi a^3(\varepsilon_m(\omega) - \varepsilon)/(\varepsilon_m(\omega) + 2\varepsilon) \tag{2}$$

where ε_m is the relative permittivity of the metal from bulk material and ω is the frequency.

In order to understand the stable nano-trapping within the heterogeneous plasmonic U-shaped trench well, we build the 2D plasmonic trapping modeling based on the Helmholtz EM equation, taking into account the gradient and scattering elements of the localized electric field. We numerically solve the modeling using the commercial software package COMSOL

Multiphysics. The localized electric field induced scattering and gradient forces on nanoscale are obtained for understanding the trapping properties within the heterogeneous U-shaped trench geometry. For the FEM simulations, here, we carefully select the FEM meshes as triangles to assure the accuracy of the simulation results [12]. Identifying the mesh best-suited for our modeling from COMSOL Multiphysics, we tried to choose the proper element type and size for U-shaped structure. The triangular mesh accuracy (e.g. the minimum length of a side of the triangular) can be reduced to 0.01 nm for the current COMSOL function, which is quite a bit smaller than the thickness of graphene sheet (0.335 nm).

In the current investigations, the U-shaped trench is immersed in water ambient medium (not shown in figure 1) via setting the index of refractive of the simulating environment, which is the usual situation of nano-manipulation for bioscience. Also, we using a plane wave for calculating the trapping properties of the heterogeneous U-trench system. The graphene with respect to the Fermi energy modifications is modeled with the surface optical conductivity that was obtained via the Kubo–Greenwood formula as follows [15]:

$$\tilde{\sigma}_{\mathrm{intra}} = \frac{2\mathrm{ie}^2 \mathrm{k_B T}}{\hbar^2 \pi \left(\omega + \mathrm{i}\tau_{\mathrm{g}}{}^{-1}\right)} \ln\left[2\cosh\left(\frac{\mathrm{E_F}}{2\mathrm{k_B T}}\right)\right] \tag{3}$$

$$\tilde{\sigma}_{\mathrm{inter}} = \frac{\mathrm{e}^2}{4\hbar}\left[\frac{1}{2} + \frac{1}{\pi}\arctan\left(\frac{\hbar\omega - 2\mathrm{E_F}}{2\mathrm{k_B T}}\right)\right] - \frac{\mathrm{e}^2}{4\hbar}\left[\frac{\mathrm{i}}{2\pi}\ln\frac{(\hbar\omega + 2\mathrm{E_F})^2}{(\hbar\omega - 2\mathrm{E_F})^2 + (2\mathrm{k_B T})^2}\right]. \tag{4}$$

In the equations, T is the temperature set as 300 K (room temperature), $\mathrm{k_B}$ is the Boltzmann constant and $\mathrm{E_F}$ is the Fermi energy of excited graphene. We only considered the intrinsic mono-layer graphene sheet.

It should be emphasized that the graphene is considered as a carbon atom sheet so that the graphene properties can be well treated as the thinned graphite permittivity [16]. Here, the U-trench 'gap/height' is considered as a variation to tune the heterogeneous plasmonic nano-trapping forces and the operating resonance wavelength. Generally, the electron beam evaporation and the electron beam lithography can be qualified for fabrication of the designed U-trench geometry. The chemical vapor deposition and the graphene transfer methods can be used to coat the graphene on the U-trench geometry [17, 18].

3. Results and discussions

The graphene-based heterogeneous plasmonic nano-trench can lead to the active tunability of the plasmonic properties, which can be hardly assured as only considering the noble gold and silver as the trapping geometry materials. In figure 2, a 0.335 nm thickness of graphene is used to cover the silver/gold/sapphire substrate, silver/silver/sapphire substrate and gold/gold/sapphire substrate U-shaped trench structures. Furthermore, we carried out the comparison of the calculated

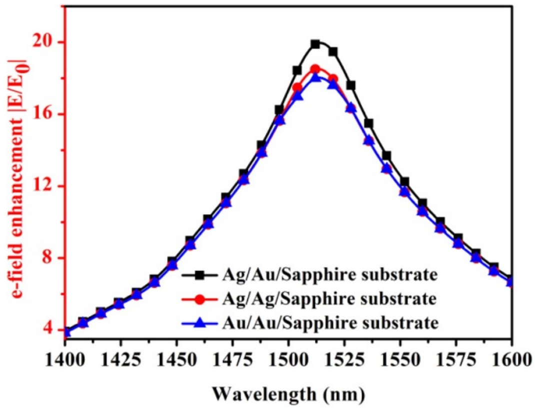

Figure 2. Calculated results of localized electrical-field (e-field) enhancement at the bottom of the 2D U-trench to understand heterogenous and homogenous U-shaped geometries. The Fermi level of graphene is $\mathrm{E_F} = 0.2$ eV. The nanosphere diameter is 50 nm here. The input power is 5.5×10^9 W m^{-2}. The gap/ height of U-trench are equal to 600 nm.

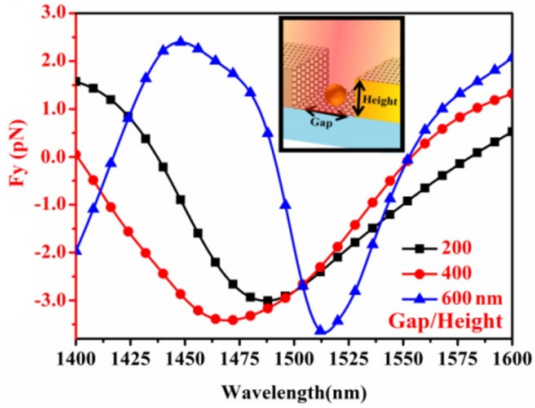

Figure 3. The vertical trapping forces in the y direction with respect to different U-trench equal gap/height sizes for the designed silver, gold and graphene heterogeneous U-trench as a function of incident wavelength. The Fermi level of graphene is $\mathrm{E_F} = 0.2$ eV. The nanosphere diameter is 50 nm here. The input power is 5.5×10^9 W m^{-2}.

e-field enhancement with homogenous geometries, such as silver/silver/sapphire substrate and gold/gold/sapphire substrate in figure 2. We can see that the e-field enhancement factor is 19.88 for silver/gold/sapphire substrate and is stronger than homogenous geometries.

The vertical trapping force in the y direction with respect to different U-trench gap/height sizes as a function of incident wavelength for the designed U-trench are shown in figure 3.

Gold sphere is considered as the target to be trapped within the designed U-trench geometry. It is clear that the calculated vertical trapping force exerting on the gold nanosphere can be typically on the scale of Pico Newton. More importantly, the vertical trapping force with respect to different U-trench gap/heights of 200 nm, 400 nm and 600 nm can be discriminatively modified by changing the incident laser wavelength. Also, we can see that the maximal vertical force of −3.75 pN appears at the wavelength of 1513 nm for the designed U-trench with 600 nm equal gap/height.

The negative value of the vertical trapping force means that the maximal vertical force is at the direction of the incident laser. It can be well understood by the gradient localized electric field generated by the surface plasmon of the designed graphene-based heterogeneous silver and gold U-trench geometry. It should be emphasized that the gold nanosphere is put 10 nm above the bottom of the U-trench geometry. In order to understand the gold nanosphere trapping stability of the U-shaped trench, we investigate the transverse trapping force in the x direction. It shows that the deepest transverse potential well in the x direction is located at the center of the gap of the U-trench due to the transverse symmetry of the U-trench geometry. As a result, the gold nanosphere will be clamped at the center ($x = 0$ nm) of U-trench. In the following investigations, we will focus on the vertical trapping force and potential well distribution in the y direction for the stable nano-trapping within the graphene plasmonic-based heterogeneous nano-trench. The vertical trapping force exerting on nanosphere made of different materials as a function of incident wavelength with respect to the nanosphere diameter is shown in figure 4. We can see from figure 4(a) that the vertical force profiles exhibit dual resonance poles at wavelengths 1448 nm (pole 1) and 1513 nm (pole 2), respectively. The vertical force at the resonance 'pole 1' keeps almost constant with decreasing the gold nanosphere diameter from 50 nm to 10 nm. However, an obvious rise of the vertical force with increasing the gold nanosphere diameter from 10 nm to 50 nm can be observed for resonance 'pole 2' at wavelength 1513 nm. It indicates that increasing the gold nanosphere diameter can lead to even stronger pushing force, causing the possible displacement of gold nanosphere toward the bottom of U-trench due to trapping force induced mechanical migration of the nanosphere. We also can see that the maximal vertical trapping force spectra have obvious red-shifts with increasing the nanosphere diameter from 10 nm to 50 nm at both of the resonance poles. It originates from the electrical polarity of gold nanosphere participating in modifying surface plasmons. A larger gold nanosphere trends to generate the multipolarity of the plasmon resonances, leading to the red-shift of the resonance wavelength. Figure 4(b) shows the vertical force as a function of incident wavelength for trapping different nanospheres made of a wide range of materials including polystyrene, silica glass, silicon nitride, sapphire, silver and gold. The Fermi level of graphene is set $E_F = 0.2$ eV and the nanosphere is put 10 nm above the bottom of U-trench. The materials are typically selected, applicable for bio-science applications like drug delivery and photothermal therapy, etc. The vertical trapping force exhibits similar tendency for all trapped sphere materials. However, the maximal vertical trapping force can be achieved by considering gold as trapping target. As a result, we focus on investigation of stable nano-trapping of typical gold nanosphere within the graphene plasmonic-based heterogeneous nano-trench, for which the gold nanospheres are also widely used for wide range of bio-applications.

The simulation results of the vertical trapping force as a function of incident wavelength with respect to the graphene Fermi energy levels are shown in figure 5. We can see that the maximal vertical trapping force can be tuned by the Fermi energy especially at the resonance wavelength of 1448 nm and 1513 nm, respectively. However, as the incident wavelength is far away from the resonance one, the Fermi energy has nothing to do with the trapping force. It can be explained as the significant participation of graphene in enhancing the localized electric field at the two distinct plasmon resonance states of wavelengths 1448 nm and 1513 nm. Interestingly, we can see that the graphene Fermi level modification to the trapping force exhibits the opposite tendency at the wavelength of 1448 nm and 1513 nm. By increasing the graphene Fermi energy from 0 eV to 1.2 eV, the trapping force in the opposite direction of laser moderately increases at resonance wavelength of 1448 nm, but the negative trapping force along the trapping laser direction is decreasing at the resonance wavelength of 1513 nm. It indicates that the trapping force for clamping the gold sphere on bottom of the U-trench can be actively tuned by graphene sheet Fermi energy, which can be modified by the static electricity tunability of graphene [10, 12]. The results can be attributed to the bonding graphene plasmon hybridization and antibonding hybridization at wavelength 1448 nm and 1513 nm for nano-trapping, respectively. The current design limit is the comparative tunability ability due to the atom layer graphene sheet bearing less electrons gas for supporting the plasmonics, which can be further enhanced as FLG is used as a substitute of mono-layer graphene.

We can see from figure 6(a) that the maximal e-field enhancement factor |E/E0| for the U-trench (equal to gap/height of 200 nm) appears at wavelength 960 nm for both situations with and without graphene. However, the maximal e-field enhancement factor for the graphene situation is 3.48 times larger compared to that without graphene. Also, figure 6(c) reveals that the maximal e-field enhancement factor for the U-trench (equal to gap/height of 600 nm) exhibits at 1513 nm for both situations graphene and without graphene. It indicates that the graphene plays an important role in the localized electrical-field enhancement at the resonance wavelength, causing the large electric-field gradient. In fact, the e-field enhancement originates from the enhanced graphene plasmon hybridization with the silver and gold of the U-trench. As a result, the trapping force can be dramatically modified as considering the graphene hybrid plasmons with the U-trench geometry.

By sweeping the angle of the incident light as shown in figure 6(b), the e-field enhancement is decreased monotonously from 8.77 to 3.07 by increasing the angle of incidence from 0° to 90°. The e-field enhancements with respect to different incident angles are calculated at the fixed resonance wavelength 960 nm. Additionally, in figure 6(d), the enhancement factor |E/E0| also decreased monotonously from 19.88 to 13.86 by increasing the angle of incidence from 0° to 90°. The results of different incident angles are calculated at the fixed 1513 nm resonance wavelength respectively. The results show that the incident angle can play a key role in affecting the e-field enhancement factor, giving rise to the potential modification of nano-trapping within the U-trench. In fact, as the linear

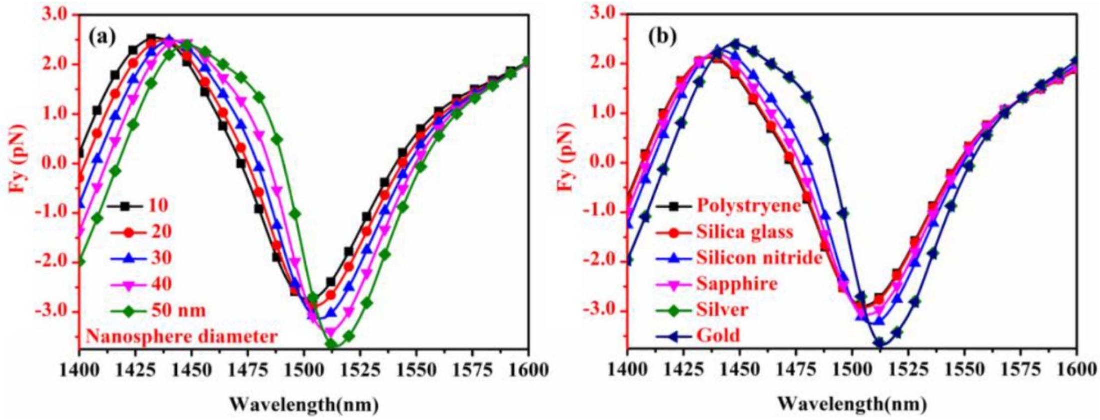

Figure 4. The vertical trapping forces as a function of incident wavelength with respect to the nanosphere diameter and elements. The Fermi level of graphene is $E_F = 0.2$ eV. (a) Considering different gold nanosphere diameter, (b) considering nanosphere made of different materials, the diameter of the nanosphere is set as 50 nm. The gap/heights of U-trench are equal to 600 nm. The input power is 5.5×10^9 W m^{-2}.

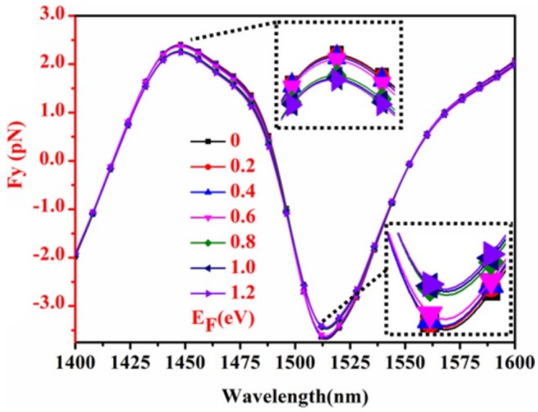

Figure 5. The simulations results of the vertical trapping forces as a function of incident wavelength with respect to the graphene Fermi energy levels. The gap/heights of U-trench are equal to 600 nm. The input power is 5.5×10^9 W m^{-2}.

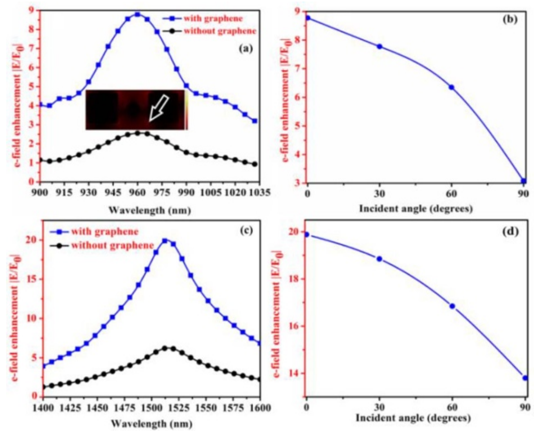

Figure 6. Calculated results of localized electrical-field (e-field) enhancement at the bottom of the 2D U-trench to understand graphene and incident angle effects. (a) e-field enhancement factor with respect to graphene and without graphene with the position of enhancement indicator. (b) e-field enhancement factor with respect to different incident angles. The input power is 3.5×10^9 W m^{-2} . 2D U-trench gap/height both are equal to 200 nm. (c) e-field enhancement factor with respect to graphene and without graphene. (d) e-field enhancement factor with respect to different incident angles. The input power is 5.5×10^9 W m^{-2}. 2D U-trench gap/height is equal to 600 nm.

polarization is considered here, the polarization can be modified with changing the incident angle. It can be attributed to the polarization-dependent plasmon polarity of the U-trench. As the laser incident angle increases, the asymmetric plasmon mode is converted to the symmetric plasmon mode, especially the completely symmetric plasmon mode can be generated as the surface-parallel polarization (incident angle at 0°) used due to the geometry symmetry along the surface of the U-trench. As the laser incident angle is at 90°, namely the laser polarization is perpendicular to the surface of the U-trench; the asymmetric plasmon mode can be excited in the U-trench.

In fact, the U-trench size also plays an important role in affecting the trapping spectrum range. As we tend to tune the trapping spectrum for typical applications at wavelengths around 960 nm to 1513 nm [19, 20], one can select the different U-trench size equal (gap/height) here as addressed in figure 7. Also, the trapping forces were investigated in the spectrum range of 1400 nm to 1600 nm, which is optimized for the specific infrared wavelength optical trapping application as reported [20].

In order to well understand the stable trapping of a nanosphere by the graphene-based heterogeneous nano-trench, we carried out the calculations of the potential well as a function of nanosphere displacement at both the vertical (y) and transverse (x) directions of the U-trench as in figure 8. Here, we focus on the 2D U-trench gap and height, both equal to 200 nm for good definition of the stable trapping of very small nanoparticles (40 nm sphere) within the U-trench. As a result, the trapping wavelength can be tuned to 920 nm. In fact, as the U-trench gap increases, a larger particle with size on the micron scale can be easily clamped in the gap of trench. In the later situation, the trapping instability becomes more inapparent due to the Brownian motion not playing a role for a

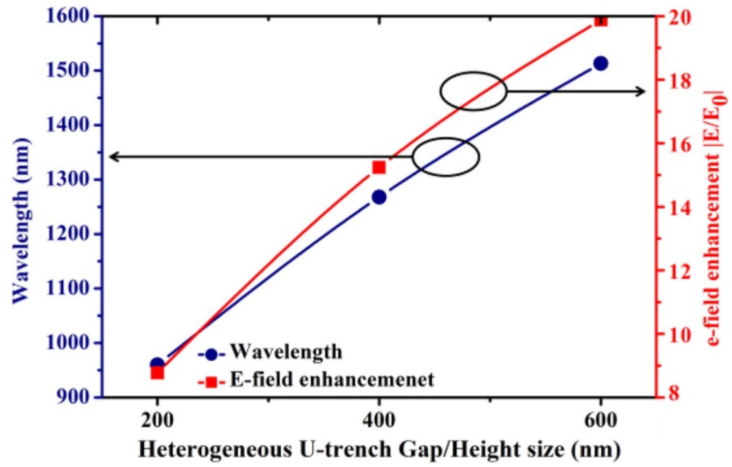

Figure 7. Spectrum range and enhancement factors originate from the different U-trench sizes.

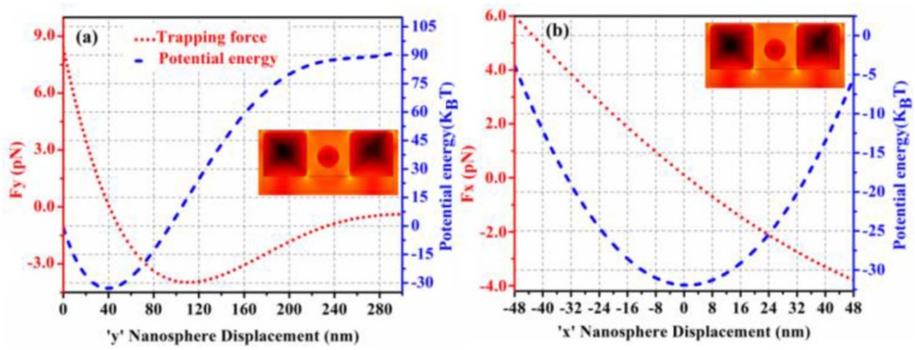

Figure 8. Trapping forces (Fy) and the corresponding potential energy (K_BT) of a 50 nm gold sphere are as a function of displacements. The input power is 3.5×10^9 W m^{-2} and the Fermi level of graphene is $E_F = 0.2$ eV. 2D U-trench equal gap/heights are 200 nm.

larger nanosphere. The trapping target is selected as the gold nanosphere based on the previously calculated largest trapping force compared to other typical spheres. We find from figure 8(a) that the deepest potential well is located at 40 nm above the bottom of the U-trench. The vertical potential well is 32 times larger than the Brownian motion energy K_BT, indicating that the nanosphere can be stably trapped within the U-trench [21, 22]. Also, the trapping forces and potential well of 32 K_BT are calculated with an incident intensity of 3.5×10^9 W m^{-2}. Moreover, our theoretical results of trapping laser intensity are lower than as previously reported, where a 50 nm polystyrene bead is dipped in water for manipulation of individual nano-units, such as viruses or large proteins [23]. It can be observed from figure 8(b) that the deepest transverse potential well is located x = 0 nm, and the transverse potential well gets 32 K_BT, indicating that the gold sphere can be stably fixed at the transverse gap center of the U-trench. As a result, the gold sphere can be stably trapped at center x = 0 nm and 40 nm above the bottom of the U-trench. Also, we can see from figure 8(a) that when the sphere moves from 0 nm to 40 nm in the y direction, the positive trapping force (Fy) exhibits a dramatic drop. However, as the sphere moves further, the trapping force changes to its opposite direction and exhibits non-monotonous modifications. Differently, the transverse trapping force experiences monotonous drop as the sphere moves from the left side (−40 nm) to the U-trench center (0 nm) and monotonous increase from 0 nm to 40 nm in the x direction (figure 8(b)). The results can be helpful for understanding the basic processes of stable trapping of gold nanosphere and potential typical spheres using the designed graphene plasmonic-based heterogeneous nano-trench.

4. Conclusion

In conclusion, we have theoretically investigated the heterogeneous plasmonic nano-trench for stable trapping of nanospheres made of noble metals and non-metals like semiconductors, polymers and dielectrics with sphere diameter from 10 nm to 50 nm. Interestingly, we observed that trapping potential can be 32 times larger than the Brownian motion energy, indicating the spheres can be highly stabilized within the heterogeneous plasmonic nano-trench. By considering the graphene plasmonic-based heterogeneous nano-trench, the gold sphere can be stably trapped 40 nm above the bottom of the U-trench bottom. The maximal e-field enhancement factor of the graphene-based U-trench is 3.48 times larger compared to that without graphene. It can be explained as significant participation of graphene in enhancing the localized electric field of the graphene-based heterogeneous nano-trench. The results can be potentially helpful for well understanding of stable trapping of gold nanosphere using the graphene plasmonic-based heterogeneous nano-structures.

Acknowledgments

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