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Stable plasmonic nano-trapping using a hybrid gold-graphene V-trench with an extremely deep potential well

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Abstract: We theoretically investigated the stable plasmonic trapping of sub-50nm targets using integrated gold-graphene V-trench with extremely deep trapping potential well. A hybrid plasmonic trapping model is self-consistently built, which considers the surface plasmons excitation for supporting the scattering and gradient optical forces on the diffraction-limit broken nano-scale. It is demonstrated that trapping of 40 nm gold nano-sphere within the designed hybrid nano-V-trench is achieved via optimal laser intensity in visible spectra. More interestingly, it is observed that the hybrid nano-V-trench forms a vertical potential well having an extremely deep potential well of 85 K_BT on the trapped 40 nm gold particle. Formation of an extremely high potential well can be explained by the perspective of localized electric field enhancement inside the nano-V-trench, which was reinforced by the involvement of graphene plasmons in the hybrid plasmonic system. This work can be helpful for well understanding of nanoparticles trapping with high stability, which is useful for the nano-manipulations in the applications of quantum dots lighting, SERS nano-sensor and nano sphere plasmonic lithography.

1. Introduction

Smart manipulation and characterizations of nanoscale particles via non-contacted optical route can be highly expected for nano-manipulations in the fields of molecule biosciences and nanosciences [1–8].Plasmonic optical trapping offers the intrinsic advantage over the conventional optical tweezers when it comes to trapping and manipulation of small targets on subwavelength size [1,2, 9,10] .Until now, much attention has been drawn in to plasmonic nanostructure assisted optical trapping due to their extraordinary capability of offering strong, sub-wavelength energy confinement while creating a high optical gradient [1,11,12].

Physically, the plasmonic trapping is supported by the resonant excitation of the plasmonic nanostructures due to the photon energy matching to that of the surface plasmons. The strong plasmonic trapping can be well-defined at the plasmon resonance state of nano-structures [2,13,14]. A wide range of plasmonic structures like nanoholes, nano-rings and nanorods have been reported for the plasmonic tweezers for the specific applications [1,15–17]. Among them, the V-shaped trenches have found huge potential for introducing robust trapping and manipulation under relatively low incident laser power and they are reliable for stable trapping due to its deeper potential wells [18–21]. Recently, it is reported that graphene as an atomic layer of graphite with a honey-comb lattice structure have found huge potential in enhancing the plasmonic field enhancement and modifying the resonance spectrum [22–26]. Our recent works also proved that not only does graphene intrinsic plasmons that are tunable and adjustable, but a hybrid

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graphene-gold V-trench promises a variety of exciting phenomena like enhancement of electric field gradients. [24,27,28].As a result, the enhanced e-field gradients can be greatly beneficial for the stable trapping due to the narrowing trapping potentials from the space localized hotspot [29]. Nevertheless, as the trapping particle goes to even smaller on molecule or quantum scales, the thermal fluctuation originating from Brownian motion still becomes dominant in deteriorating the trapping stability [2,9,10,30]. An alternative method to overcoming the Brownian motion is to increase the laser power of the trapping geometry. In fact, high optical power trapping leads to possible optical damage of the trapped specimen and this could highly subjective when trapping and manipulating biological cells or flexible electronic devices [9,31]. It is still a challenging for exploring the highly stable plasmonic nano-trapping on sub-50 nm scale from both theoretical and practical aspects due to the potential interest in stably arranging the nanodots for applications in the fields of quantum dots lighting, SERS nano-sensor and nano sphere plasmonic lithography etc. [32–35].

In this paper, we theoretically investigated the stable trapping of 40 nm gold particle and flexible manipulation of other sized nanoparticles made of engineering materials from metal to semiconductor by designing the integrated gold-graphene plasmonic V-trench. The trapping forces and potential well within the V-trench are modeled and numerically analyzed in details using finite element method (FEM). By considering the hybrid nano-V-trench architecture composed of gold and graphene, we have shown the stable optical trapping of 40 nm gold nanoparticle immersed in water, which is well achieved with the peak potential well depths of 85 K_BT. Formation of extremely high potential well is explained as the localized electric field enhancement inside the graphene hybrid nano-V-trench.

2. Structure design and modelling

Proposed gold- graphene integrated hybrid plasmonic nano V-trench optical trap is illustrated in Fig. 1. A 2D plasmonic simulation model which is depicted in Fig. 1 (b) is designed and investigated to study the plasmonic optical trapping characteristics and stable optical trapping. The 240 nm wide plasmonic nano V-trench optical trap having a 70° slope is fabricated out of a 350 nm thin gold (Au) film which is deposited on a monolayer (0.335 nm thin) of graphene. The entire hybrid plasmonic V-trench optical trap nanostructure is fabricated on a silica (SiO_2) substrate. The geometry of the optical trap is kept a constant throughout the investigation. Whole nanostructure is immersed in liquid which serves as the medium for trapping particles and the V-trench geometry is irradiated with a focused laser beam vertically from above as shown in the Fig. 1(a) such that the polarization direction of light is perpendicular to the plane of incident light which matches the momentum of plasmons. A perfect matching layer (PML) is defined surrounding the whole nano trench geometry and the immersed liquid environment.

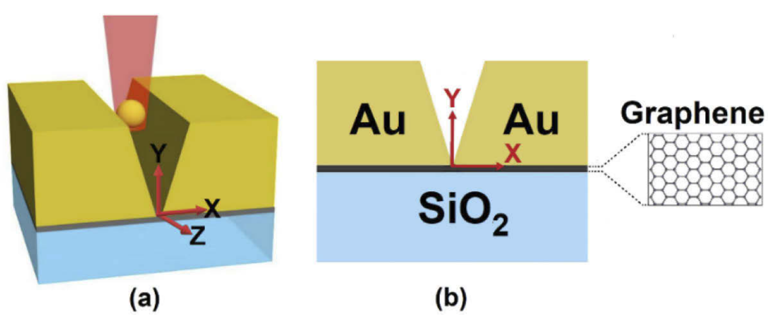

Fig. 1. (a) 3D schematic of the hybrid gold-graphene V-trench plasmonic nanotrap with gold sphere inside the trench being trapped with trapping laser from the top, (b) 2D simulation schematic of the hybrid gold-graphene V-trench plasmonic nanotrap.

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Triangular FEM mesh with definite nano-scale geometry has been selected to carryout accurate FEM process during the simulation. Near-field scattering phenomena in the nano-trench is modeled by the Helmholtz electromagnetic equation where we obtained the numerical solutions of the Helmholtz equation via solving the built matrix. Analytical dielectric function of gold and other materials used in the simulation model are extracted from the experimental datum via numerical fitting [36]. 3D schematic of the proposed hybrid plasmonic nano V-trench optical trap is illustrated in Fig. 1 (a). Chemical vapor deposition (CVD) technique and the graphene transfer methods is used to coat a monolayer of graphene on the silica (SiO_2) substrate. Furthermore, using electron beam evaporation technique, a gold film is coated on top of the graphene layer where the nano V-trench is milled on the gold film using a focused ion-beam (FIB) milling technique [37,38].

The surface plasmon can be resonantly excited depending on the material used in the plasmonic structure, shape and sizes of the structure, incident laser wavelength and the effects of interactions between particles in the ensemble. Plasmonic coupling causes the localized electric field enhancement, which augments the trapping force. But unlike non-graphene plasmonic optical tweezers, localized field enhancement can be flexible-tuned by finely altering the Fermi level of the graphene layer. Fermi energy (EF) of graphene is given by [39];

$$\mathrm{E_F} = \hbar \mathrm{v_F k_F} = \hbar \mathbf{v_F} \sqrt{\pi \mathrm{n_{g,2D}}} \tag{1}$$

where $\hbar$= Modified Planck's constant = 6.582×10^{-16} eV.s, $\mathrm{v_F}$ = Fermi velocity $\approx 1\times 10^6$ m/s, $\mathrm{k_F}$ = Fermi wave vector, $\mathrm{n_{g,2D}}$= carrier concentration in two-dimensional graphene sheet.

Rayleigh approximation has been taken in to account throughout the investigation for all trapped particles which are smaller than the wavelength of the incident trapping laser. Plasmonicly enhanced local electric field is confined within the near-field of the trap generating a considerably strong field gradients and large local field intensities, in return augmenting optical forces acting on the trapped particle to create a strong stable optical trap. Optical forces are calculated based on the dipole approximation which can be determined as follows by considering a nanosphere of radius "a" [40,41];

$$\boldsymbol{F} = \frac{1}{4}\varepsilon_0 Re\{\alpha_0\}\nabla|\boldsymbol{E}|^2 + \frac{n\sigma}{2c}\{\boldsymbol{E}\times\boldsymbol{H}^*\} + \frac{\sigma}{2}Re\left\{i\frac{\varepsilon_0}{\boldsymbol{k}_0}(\boldsymbol{E}.\nabla)\boldsymbol{E}^*\right\} \tag{2}$$

where ε_0 = Vacuum dielectric permittivity, α_0 = Polarizability of a point-like particle, E = Incident electric field, n = Refractive index of surrounding medium, σ = Total cross-section of the particle = k Im (α_0), c = Speed of light, H = Incident magnetic field, k_0 = Wave vector in free space.

The trapping potential resulting from the optical forces is a key factor that determines the stability of the optical trap, and it can be obtained by [9];

$$U(\boldsymbol{r}_0) = \int_{\infty}^{r_0} \boldsymbol{F}(\boldsymbol{r}).dr \tag{3}$$

FEM with a fine triangular mesh was used in the core numerical analysis for the investigation of plasmonic optical trapping characteristics. The interaction between the particle trap system and the trapping laser leads the way to the generation of optical forces on the particle. Under the investigation we focused on local electric field behavior inside the trench and trapping force characteristics, trapping potential characteristics and trapping force variations for different immersed mediums. Furthermore it can be pointed out that, wide range of nanospheres can be well manipulated while the proposed hybrid gold-graphene V-trench plasmonic nanotrap being immersed in water and specifically 40 nm gold nanosphere can be stably trapped within the geometry.

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3. Results and discussion

As shown in Fig. 2 electric field distribution color maps inside hybrid plasmonic V-trench optical trap with 40 nm gold nanoparticle which is being manipulated at 280 nm trap depth with different incident wavelengths, an evident localized electric field enhancement can be observed at resonance wavelength. Color-map representing the resonance condition undoubtedly demonstrates a local electric field enhancement inside the graphene based nano-V-trench. Localized field enhancement can apparently be characterized as optical bondage in the nanogap [42]. Incident light with the resonance wavelength causes the excitation of localized surface plasmons inside the hybrid nano-V-trench| particle system where a strong light scattering occurs in an appearance of a local electromagnetic field enhancement [43].

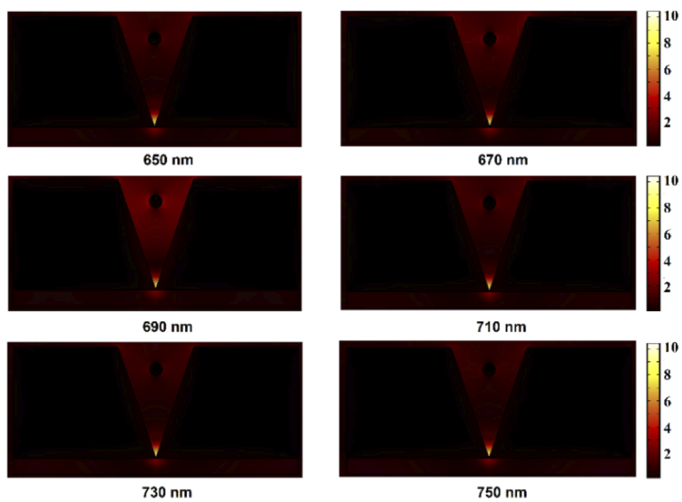

Fig. 2. Electric field distribution color maps inside the nanotrench while 40 nm gold nanoparticle being kept at y = 280 nm position with respect to variation in the wavelength of the incident light beam.

When the Localized Surface Plasmon Resonance (LSPR) condition is satisfied, the electric fields in the vicinity of the particle's surface are greatly enhanced [16] and moreover in the presence of visible light irradiation the gap-mode excitation comes in to the action at the resonance wavelength which leads to gap-mode transitions resulting remarkable enhancement in the electromagnetic field of the incident light inside the trapping nano-V-trench. At 690 nm incident wavelength a clear localized electric field enhancement can be seen. The electric field spatial distribution inside the V-trench region and the region near to the surface of the trapped 40 nm gold-particle are strongly concentrated. These regions where stronger electric field enhancements exist are often known as hot spots. Previous studies have proven that metallic nanostructures such as nanogap, nano-junction, nanoantenna, nanobowtie, can act as small light sources in the nanospace [42]. Gap-mode field localization supported electric field gradient inside the V trench can be significantly enhanced irrespective of the diffraction-limit at nanoscale, which is greatly beneficial for nanoscale optical trapping.

In comparison to a homogeneous gold (Au) nano-V-trench plasmonic optical trap with same dimensions and irradiation conditions, proposed hybrid gold-graphene V-trench trap exhibits an obvious augmentation in local electric field enhancement ($|E/E_0|$) inside the V-trench (Fig. 3) . An electric field enhancement of more than 3.5 times can be clearly observed for the graphene based nano-V-trench optical trap in visible spectral range from 630 nm to 700 nm. At 690 nm both field enhancements reach their maximum values, 5.25 for graphene integrated hybrid plasmonic

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optical tweezer and 4.625 for homogeneous gold plasmonic optical tweezer respectively. It is a 13.6% local electric field enhancement in hybrid gold-graphene V-trench with respect to the homogeneous gold V-trench based plasmonic optical tweezer. It should be mentioned that the Fermi level of the graphene monolayer of hybrid gold-graphene V-trench is constantly maintained at 0.2 eV during the field enhancement comparison in the given spectra. Significant electric field enhancement is caused due to the presence of graphene plasmons which alters the plasmonic environment of the hybrid gold-graphene V-trench optical tweezer.

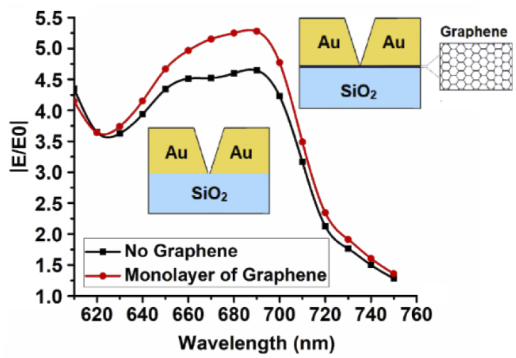

Fig. 3. Comparison of local electric field enhancement ($|E/E_0|$) inside the nano-V-trench between gold (Au) based plasmonic nano-V-trench optical tweezer and gold and graphene based hybrid plasmonic nano-V-trench optical tweezer with monolayer of graphene (thickness 0.335nm). Laser power intensity = 4×10^9 W/m^2, immersed medium = water (refractive index (n) = 1.33).

Graphene with its high intrinsic electron mobility which can be high as 200,000 cm^2/Vs adds an extra boost to the total plasmonic field inside the nano-V-trench. Furthermore it is a well-known fact that electron mobility of graphene can be altered by changing the Fermi level which could be useful to achieve tunable plasmonic environment for nanoscale optical traps [23]. The vertical trapping force (F_y) as a function of the vertical (y-axis) displacement locus of a 40 nm gold particle immersed in oil with respect to different graphene Fermi levels is illustrated in Fig. 4 (a). Maximum trapping forces can be achieved at 237 nm above the bottom of the V-trench by tuning the Fermi level at resonance wavelength 670 nm. More importantly, it can be observed that alteration in Graphene Fermi energy leads to changes in vertical trapping forces inside the V-trench. This clearly indicates that trapping force for manipulating the 40 nm gold sphere inside the trench can be actively tuned by alteration in Fermi energy of monolayer graphene which can be made possible with electrostatic gating of graphene [24,25]. Tunability of the vertical trapping force exhibits the remarkable ability of graphene plasmons to alter the local electric field inside the trap. Changes of the Fermi energy would lead to a simultaneous change in the interband threshold and the interband transition owing to the unique linear dispersion relation of graphene. As a result, tunable Fermi energy leads to tunable surface optical conductivity, which includes the contributions from intraband transitions and interband transitions of excited graphene [44]. This phenomena in the nano V-trench optical trap can be attributed as the plasmonic hybridization in to the total plasmonic environment by graphene and gold plasmons. It occurs with the coupling between charge carriers in monolayer of graphene and gold V-trench which results in LSPR that is considerably stronger than the LSPR of homogeneous gold nano V-trench of same dimension [45].Fig. 4 (b) illustrates the vertical trapping force acting on the particle as a function of trapping particle size with respect to different types of nanoparticles. Plasmonic coupling associated between the particle and the optical trap, deviates based on the particle size and particle refractive index. Strongest trapping force can be predicted for a 38 nm size gold nanoparticle with the

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magnitude of 9.2 pN. Compared to dielectric nanoparticles, gold exhibits strongest plasmonic coupling due to its high conductivity and polarizability and influence of LSP enhanced local electric-field in the vicinity of the particle [2].

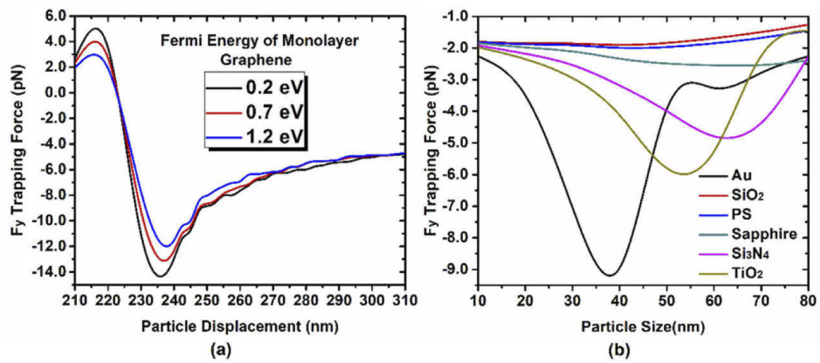

Fig. 4. (a) Vertical trapping force as a function of particle's vertical displacement locus (y-axis) with respect to three different Fermi energy levels of the monolayer graphene (thickness = 0.335 nm). Immersed medium: oil (n=1.518). Laser power intensity = 4×10^9 W/m^2. Particle = 40 nm gold nanoparticle. (b) Evolution of the vertical trapping force F_y for particles made of different materials (Gold (Au), silica glass (SiO_2; n = 1.54), polystyrene (PS; n = 1.59), Sapphire (n = 1.77), silicon nitride (Si_3N_4; n = 2.2) and titanium dioxide (TiO_2; n = 2.7)) as a function of particle size. Particle is kept stationary at 280 nm vertical position while being immersed in water and irradiated by 4×10^9 W/m^2 incident laser power at 670 nm fixed wavelength

In the other hand, strong trapping forces acting on the particle are associated with the resonance condition within the plasmonic optical trap. Resonance is determined by absorption and scattering which are dependent on the shape, size and the dielectric constant of the trapped particle and plasmonic trap itself. Surface geometry of the trapped nanoparticle changes with the change in the shape or size of it. The change in surface geometry causes a shift in the electric field density on the particle surface which in return causes a change in the oscillation frequency of the electrons, generating different cross-sections for the optical properties including absorption and scattering [46]. Figure 5 shows trapping force acting on 40 nm gold particle in different immersed mediums (water (n = 1.33), blood plasma (n = 1.351), ethanol (n = 1.361), formamide (n = 1.428), glycerin (n = 1.473) to oil (n = 1.518)). The vertical trapping force spectra of gold nano particle in all surrounding immersed mediums have valley that indicates maximum trapping force. The maximum trapping force position of the 40 nm gold nano particle is found to be redshift as the surrounding refractive index of the immerse medium increases. As the refractive index increases from 1.33 (water) to 1.518 (oil) the maximum vertical trapping force on the particle gradually increase from 16.01 pN to 20.6 pN. This fascinating phenomena can be well explained by focusing on the factors that determines LSPR. For a single plasmonic nanoparticle, the localized plasmonic peak position, absorption and scattering yields and the plasmon field intensity and distribution are determined by its shape, size, material, and dielectric constant of the surrounding immerse medium.

When the immerse medium (dielectric medium) inside the hybrid gold-graphene V-trench is changed, the plasmon field is distorted, and the plasmonic absorption and scattering spectra could shift differently. In other words LSPR frequency can be shifted through alteration of surrounding medium. These shifts are basically due to the electromagnetic interactions between the localized modes which are generally of a dipolar nature [47], [48]. The red-shift of the maximum trapping force with increasing refractive index of the immersed medium is caused due to the coupling of

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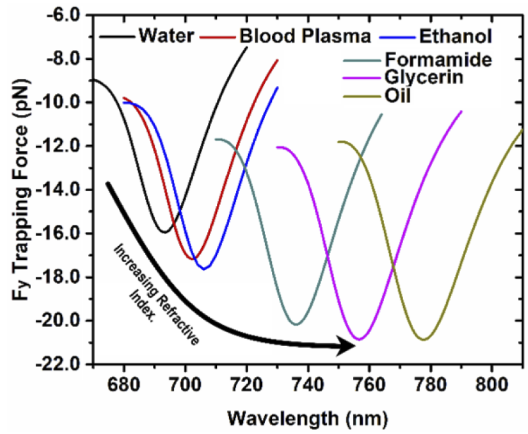

Fig. 5. Vertical trapping force acting on a 40 nm gold particle as a function of incident trapping laser spectra for different surrounding mediums of the optical trap; Water n=1.33, Blood Plasma n=1.351, Ethanol n = 1.361, Formamide n = 1.428, Glycerin n = 1.473 and Oil n = 1.518. Trapping laser power intensity 4×10^9 W/m^2.

the localized surface plasmon with the polarization dipoles in the dielectric surrounding medium inside the nano-V-trench. Higher refractive index dielectric materials have higher electric dipole polarization density, therefore resulting in stronger coupling to surface plasmons with red-shift in the incident laser spectra [49]. Many of potential applications of the tunable force via different environment mediums lies in the fields of microbiology and genetic engineering. For an example the test liquid Formamide used in this experiment is mostly used in ds-DNA manipulation. So tunable force via different environment in the proposed optical trap can be employed to compare interaction kinetics between ds-DNA and Formamide and other liquids [50].

To better illustrate the stable trapping capability of an extremely small target particle with proposed hybrid gold-graphene V-trench plasmonic optical trap, it is essential to demonstrate the existence of an equilibrium position at 2D space with deep trapping potential well. Figure 6 illustrates trapping force acting on a 40 nm gold particle and corresponding potential well in both x and y directions inside the V-trench. Trapping force and potential have been calculated considering locus of the target particle inside the 2D space of the optical trap. Figure 6 (a) shows the vertical trapping force acting on the particle at different positions of its locus in the y-axis and the corresponding trapping potential-well. Related displacement locus is shown in Fig. 6 (b). It is noted that all the trapping forces and potential wells are calculated with 4×10^9 W/m^2 laser power intensity at 670 nm incident wavelength. Results show that 40 nm gold particle can be stably trapped at 260 nm above the bottom of the V-trench where the trapping potential reaches its maximum potential well depth of 85 K_BT (where K_B is the Boltzmann's constant, and T = 300 °K which is the trapping temperature). Generally an optical trap with potential depth larger than 10 K_BT can be considered as stable [2,9]. Such deep potential wells are essential to damp the Brownian motion induced by the liquid immerse medium inside the trap [2,9]. It should be noted that similarly high potential-well depths had been recorded with other literatures for stable optical trapping [47]. Moreover at 260 nm y-position a clear vertical equilibrium can be seen on the trapped particle as there's no vertical trapping force (F_y=0) acting on it. The transverse trapping force acting on the particle at different positions of its locus in the x-axis and the corresponding trapping potential-well are shown in Fig. 6 (c). It can be observed that the deepest potential well for the particle moving along the x-axis reaches at x=0 with a potential depth of 48 K_BT which also exceeds the 10 K_BT stable trapping margin [2,9].Strong trapping forces in piconewton range and extremely deep potential wells are caused by plasmonic hybridization effect, which

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occurs between the Gap-Surface Plasmons (GSPs) that are confined between the sidewalls of the gold-graphene integrated V-trench and the Localized Surface Plasmons (LSPs) confined at the bottom of the V-trench and LSPs on the trapped 40 nm gold particle. This leads to hybridization enhanced electric fields inside the nano V-trench optical trap and the narrow gap between the trapped particle and the V-trench, which in return induces strong trapping forces and extremely deep potential well on the trapped 40 nm gold particle [51].Furthermore 40 nm gold particle is at equilibrium state with no transverse trapping force ($F_x = 0$) when it reaches to the deepest potential-well at x=0. With the results obtained from both Fig. 6 (a) and Fig. 6 (c), it can be clearly concluded that the hybrid gold-graphene V-trench can perform a stable optical trapping ($F_x = 0$, $F_y = 0$ and potential well deeper than 10 $K_B T$) on a 40 nm gold particle at an equilibrium position located at the center of the transverse direction (x=0) 260 nm above the bottom of the V-trench. And more interestingly, incident laser intensity (4×10^9 W/m^2) used to demonstrate stable trapping in this research is several orders lower than intensities used in conventional optical traps and some plasmonic optical traps [1,52], which would be a plus point in trapping soft target particles vulnerable to laser induced heating.

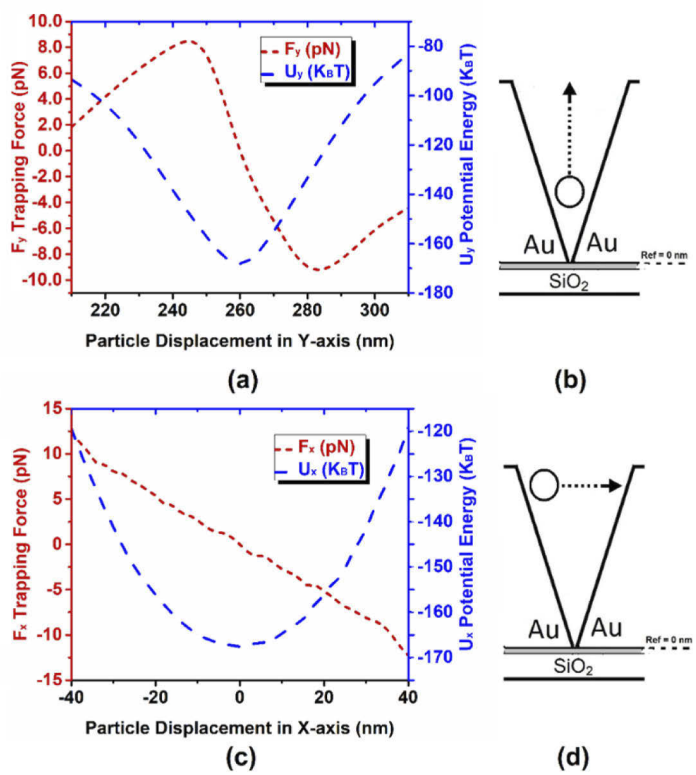

Fig. 6. (a): The vertical trapping force exerted on the 40 nm gold particle and the potential energy distribution as a function particle's displacement locus along the y-axis of the hybrid gold-graphene V-trench optical trap working at 670 nm wavelength. (b): Illustration of the displacement locus of the particle along y-axis. (c) The transverse trapping force exerted on the particle and the potential energy distribution as a function particle's displacement locus along the x-axis of the V-trench optical trap. (d): Illustration of the displacement locus of the particle along x-axis. (Input power intensity is 4×10^9 W/m^2, nanoparticle is immersed in water and Fermi level of graphene monolayer is 0.2 eV.)

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4. Conclusion

In summary, we propose a novel hybrid gold-graphene V-trench plasmonic optical trap that is capable of stable manipulating different types of sub-50 nm nanoparticles by hybridization of graphene with gold nano-V-trench, we observed a 13.6% increase in electric field enhancement with respect to homogeneous gold nano-V-trench. It is revealed that 40 nm gold particle was stably trapped in an equilibrium position inside gold-graphene V-trench with extremely large potential-well depth (85 K_BT) under relatively lower laser power intensity. Tunability of trapping forces was also achieved by varying the Fermi level graphene layer. The proposed hybrid gold-graphene V-trench plasmonic optical trap extremely deep potential well would be promising for integration of practical applications such as lab-on-chip devices for manipulation and analysis of biological entities, such as vesicles and viruses. .

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Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

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