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Graphene-based plasmonic tweezers

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ABSTRACT

Conventional plasmonic tweezers with the ability to attract and immobilize nearby sub-diffraction limit sized particles can only enhance the trapping efficiency by changing the shape of the metal nano-structures. There are several problems with conventional plasmonic tweezers. First, trapped particles can easily escape from the trap by disturbances coming from the heat absorption of the metallic surfaces. These disturbances prevent prolonged observation of the trapped particles. Second, observation of the particles becomes a challenge because the opaqueness of the metal blocks the illumination pathways. These problems can be solved by using graphene, which has high transmittance and thermal conductivity. The carrier density of the graphene is tuned by externally controlling the Fermi level through the gate voltage. Tuning the carrier density alters the local field enhancement factor far beyond the capabilities provided by other metal-based plasmonic structures. In this paper, we have shown that particles can be trapped by graphene nanoholes with larger forces than gold nanoholes. The trapping forces on gold and graphene nanoholes were compared to illustrate the benefit of graphene nanoholes. Furthermore, various trapping modes of a particle under various geometries and configurations of graphene nanoholes is discussed.

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1. Introduction

Optical tweezers allow non-invasive fine positioning and orientation control of microscopic objects [1]. Unlike other directcontact manipulation tools, such as AFM (atomic force microscopy), optical tweezers are suited for manipulating microbiological organisms, and this fact has been shown in several reports published in the field of biology and medicine [2-6]. Unfortunately, the lower size limit of target biomolecules is somewhat bounded by the diffraction limit of a laser spot, which is proportional to the wavelength of the incident beam. The reason behind this limitation is as follows. As particles get smaller, there are two main effects that act against a stable trap. First, the magnitude of the restoring force drops proportionally to the third power of the radius, which results in a reduced size of the trapping volume. Second, the viscous drag in the surrounding medium that dampens the Brownian motion also decreases. These two effects adversely influence stable trapping [7]. The optical force can be increased by increasing the incident power, but the downside is that the particle can be inflicted with thermal damage. Until the recent introduction of plasmonic

* Corresponding author. E-mail address: lygu@gist.ac.kr (Y.-G. Lee). trapping, most optical trapping has been limited to sizes above the diffraction limit [8].

Plasmonic trapping is a technique based on extreme electromagnetic field enhancements at the metal-dielectric interface of geometric features exhibiting localized high curvatures [9-11]. Examples include holes drilled on a metal-film substrate and metallic-pillars extruding from a flat substrate. Electromagnetic waves coupled with the free-electrons at the metal-dielectric interface produce extraordinary field enhancements, and nearby particles get pulled towards the high field concentration spots. Gold is the most often used metal in plasmonic trapping [12-25]. The enhancement depends on the geometry of the features and is broadly classified into two types: nanoholes [12-16] and nanopillars [17-25]. Nanopillars typically show stronger trapping stiffness [12]. However, nanopillars exhibit one major drawback. The protruding features in nanopillars behave as thermally insulated islands, and heat generated through the absorption of the incident beam is gradually accumulated, raising the temperature of the surrounding medium to the liquid boiling point. Although nanoholes exhibit better heat dissipation through conduction, they do not entirely solve the concentrated heat generation at high laser power, limiting the upper bound of applicable optical forces. Both types occlude the trapped particle or the illumination pathways such that observation becomes a challenge.

These problems, impaired observation and the limitation in the optical forces due to heat accumulation can be improved through the use of graphene in place of metals. Graphene is a one-atomthick (0.34 nm) layer that forms a 2D honeycomb structure [26]. Graphene is transparent to light in a broad spectrum, and the energy absorption is only 2.3–8.7% [27,28]. Because it is a thin film that is transparent to illumination, graphene can be placed in any location between the observer and the illuminator without affecting the imaging. The extremely low energy absorption of 2.3% in the NIR (near-infrared) spectrum means extremely high laser power can be used without causing cavitation of the liquid medium [28]. Furthermore, even the slightest heat absorbed from the incident beam can be quickly removed due to the high thermal conductivity, which is approximately 5000 W/m K [29], much higher than that of gold (318 W/m K) and copper (408 W/m K) [30].

Graphene exhibits extremely high intrinsic electron mobility of $200,000 \text{ cm}^2/\text{V s}$ [31,32]. This carrier density can be tuned by the Fermi level, controlled externally through the gate voltage [33,34]. By dynamically controlling the gate voltage, available free electrons can be finely tuned. In contrast to graphene, metals have fixed free electrons. An important consequence of this dynamic nature of graphene is as follows. The field enhancement can be varied by controlling the conductivity of the graphene for a fixed incident laser intensity. Furthermore, it has been calculated theoretically that unprecedented high field enhancement can be achieved using graphene as a free-electron carrier [35–43]. For example, Zhu et al. showed numerically that a particle can be trapped with extremely high trapping stiffness using coupled graphene strip waveguides that are separated by a 10 nm gap [44]. The downside of their method is that the geometry of the gap only allows the particles to be trapped when they are small enough to enter this gap, which necessitates additional flowing methods to drive the particles towards the narrow gap.

In this paper, using the unique features of graphene, we present a novel structure that exhibits enhanced trapping forces. The structure is based on a thin graphene layer patterned with holes, and it does not have the abovementioned geometric hindrance problem inherent in graphene strips. Graphene is used as an electron supporting layer instead of the commonly used metals in plasmonic trapping, resulting in higher field enhancement. Additionally, the proposed novel structure alleviates the observation impairment and heat generation problems often found in conventional metal-based plasmonic trapping.

2. Numerical setup

Fig. 1 illustrates the graphical modeling of the numerical analysis we performed using the finite-difference time-domain (FDTD) method. In both cases shown above, we have an identical polystyrene sphere sitting at the rim of a circular hole fabricated on a gold (Fig. 1(a)) or a graphene (Fig. 1(b)) layer. Both are suspended above a glass (SiO₂) substrate. The incident laser is considered as a planar wave, which is justifiable even for a diffraction-limited, focused, Gaussian beam due to its relatively small spot diameter (300 nm) compared with the wavelength of the beam (1064 nm). The beam is polarized in the x-axis and is denoted as p-polarized in both figures. The intensity of the laser beam is set to 0.181 mW/ μm². The incident beam is introduced in the simulation domain through the total-field scattered-field (TFSF) method. In the FDTD solver, the frequency-dependent dielectric constant of the gold is derived from the work by Johnson and Christy [45] and that of the single graphene layer is accredited to Hanson [46]. The solver leapfrogs in time and space until steady-state behavior of the electromagnetic field is reached. Subsequently, a six-sided box is monitored, encompassing the subject of interest (polystyrene particle), and the Maxwell stress tensor (MST) is summed to obtain the force that the particle is experiencing at the particular location near the rim of the hole. To prevent the six-sided box from touching the particle surface or the substrate, the particle is distanced by four grid spaces from the top gold or graphene surface. The box is depicted as a square in both figures.

A full, 3-dimensional analysis of a graphene substrate requires accurate modeling of the thickness, which necessitates a mesh grid size that is smaller than the thickness of graphene, which is only 0.34 nm. A mesh this small was found to be impractical in terms of the required amount of computation time. For this reason, the numerical model we used is based on a 2-dimensional approximation of the graphene layer. We used the commercially available FDTD solutions from Lumerical Solutions, Inc. Using this software, a graphene layer was modeled as a 2-dimensional surface conductivity and was simulated. The 2-dimensional surface conductivity approach uses the Dyadic Green's function, and we refer to the proper citation for the detailed methods [46]. This simplified model reduces the required calculation time by 90% compared using a full, 3-dimensional, volumetric permittivity approach [47].

3. Mechanism of graphene-based plasmonic tweezers

When light is incident on a metal dielectric interface, the resulting electric field on the surface, called the surface plasmon, exhibits far greater magnitude than the incident electric field. The ratio between the two is determined by the permittivity of the metal and the dielectric medium, as shown in Eq. (1) below, adapted from the work of Weber et al. [48].

$$\frac{\left|E_{SP}\right|^{2}}{\left|E_{0}\right|^{2}} \cong \frac{2\left(-\varepsilon_{2}'\right)^{1/2}\left(-\varepsilon_{2}'-\varepsilon_{1}\right)}{\varepsilon_{1}^{1/2}\varepsilon_{2}''},\tag{1}$$

In Eq. (1), the following notations were used. E_0 is the electric field magnitude of the incident light, ESP is the electric field magnitude of the surface plasmon, ε_1 is the permittivity of the dielectric medium, ε_2 is the real part permittivity of the metal, and $\varepsilon_2^{"}$ is the imaginary part permittivity of the metal. When the wavelength of the incident light is fixed, the ratio shown in Eq. (1) is determined by the permittivities of the metal and the dielectric medium. The real part permittivity of the metal is proportional to the number of free electrons, and this value is also directly proportional to the ratio in Eq. (1). Note that the real part permittivity is constant at a fixed wavelength for a metal, but it is tunable for a graphene because, in the case of a graphene, the free electrons can be increased by tuning the carrier density. In other words, graphene has an advantage compared with the metal because the real part of the permittivity can be increased, even at a fixed wavelength of incident light. Thus, the use of graphene enables greater control of the field enhancement.

4. Electric field distribution of two materials

We first performed the simulation without the particle present. The permittivity of gold depends on the incident wavelength. The optical properties of graphene depend on the Fermi level, which influences the carrier density and in turn changes the amount of available electrons. We conducted a numerical experiment to maximize the field enhancement. A 300 nm nanohole was drilled into the gold and graphene substrates and illuminated with a unit (1 V/m) length electric field. Fig. 2 shows the electric field distribution of the gold and graphene nanoholes by emitting a laser onto them

To thoroughly investigate the effect of the graphene nanohole

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