Optofluidic control using plasmonic TiN bowtie nanoantenna [Invited]

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Abstract: The performance of plasmonic titanium nitride (TiN) nanoantennas for the manipulation of fluidic flow and suspended particles in an optofluidic chip is studied. A unified theoretical framework is utilized to model the multidisciplinary problem that comprises optics, thermodynamics, and hydrodynamics. Using multiphysics finite element analysis, we simulate the temperature rise resulting from the photothermal heating of a plasmonic TiN bowtie nanoantenna (BNA) and the accompanying hydrodynamic flow generated in a microfluidic channel. We show that the TiN BNA enables over three times higher electrothermoplasmonic flow velocity in comparison to a gold BNA under similar signal conditions. Our analysis shows that TiN BNAs at near-IR biological transparency wavelengths can be utilized to initiate strong microfluidic flow for directed transport and trapping of target nanoscale objects. This makes TiN an excellent plasmonic material choice for optically controlling heat, fluidic dynamics and heat-induced forces in microfluidic devices.

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1. Introduction

Plasmonic nanoantennas provide unprecedented capability to confine light to deeply subwavelength scales and to enhance the local light field intensity [1–3]. This local electromagnetic field enhancement and confinement is enabled by the coupling of the delocalized electron cloud in metals with the incident photons to generate surface plasmons. The surface plasmon wave provides the possibility to enhance light-matter interaction for myriad applications in sensing [4], imaging [5], quantum photonics [6,7] and information processing. A key emerging application of resonant plasmonic nanoantennas in optofluidics is for enhanced optical trapping of nanometric objects. Optical tweezer technology has emerged as a powerful technique to manipulate microscale objects such as bacteria, colloidal particles and cells [8–10], and was recently recognized with a Nobel Prize in physics awarded to Arthur Ashkin for pioneering the development of optical tweezers. In the conventional optical tweezers, the diffraction limit precludes the low power trapping of nanoscale objects [11]. Since plasmonic nanoantennas can confine light to nanoscale volumes, they have generated significant interest as a platform for generating the tight trapping potential wells needed to trap nanoscale objects [11,12]. Different kinds of nanoantenna geometries have been explored including nanopillar [13,14], coaxial aperture [15–17], double-nanoholes [18–20], bowtie aperture [21], dimers [22], and circular nanoholes supporting self-induced back-action [23]. Furthermore, in addition to an enhanced local light field, the plasmonic nanoantennas also efficiently absorb light and dissipate heat, thereby providing the means to engineer the thermal landscape at the nanoscale [24–26]. This heating may be leveraged to induce thermophoretic forces [27–31] and microfluidic flows [32,33] to enable the manipulation of suspended objects. The introduction of electrothermoplasmonic tweezers [34–36] resulted in
plasmonic tweezers with a large radius of action, to overcome the diffusion-limited transport of objects while permitting the high-resolution trapping of nanoscale objects. This finding is now being employed for applications in biological sensing and directed assembly [37,38].

Traditionally, gold has been used as the material of choice for designing and experimentally demonstrating plasmonic optical tweezers in an optofluidic system due to its good optical properties and chemical stability. Advances in plasmonic materials research have ushered in alternative materials with optical properties across a broad range of the electromagnetic spectrum from UV to NIR such as aluminum [39–43] and transition metal nitrides [44–48]. Transition metal nitrides, particularly titanium nitride (TiN) has attracted significant attention because of their good plasmonic properties and photothermal response in the near-infrared biological transparency window. The interaction of the plasmonic nanostructures with light in a fluidic media is a complex process that involves several physical mechanisms including the interplay between resonant light absorption, heat generation and transport, perturbation of the thermophysical and electrical properties of the fluid, and the generation of fluidic motion. Furthermore, the application of an AC electric field to a fluidic medium with inhomogeneous dielectric properties resulting from a temperature gradient gives rise to additional volumetric body forces namely electrothermal forces to induce electrothermoplasmonic (ETP) flow in the fluid. The interplay of the multiphysical mechanisms described above can be understood via a Multiphysics modeling of the respective differential equations describing those physical effects.

In this article, we present theoretical results characterizing the performance of TiN as a plasmonic material for application in plasmon-enhanced optofluidic control. By using Multiphysics modeling, we investigate the electromagnetic, photothermal and accompanying fluid dynamics induced by TiN BNA and Au BNA submerged in a microfluidic chip. The coupled electromagnetic, heat transfer and fluid dynamics problem is solved numerically using COMSOL Multiphysics (v 5.3a) simulation software, and this analysis builds on our previous work reported in [34]. In contrast to earlier models of electrothermal flow that have considered only joule heating in the fluid induced by an applied AC electric field, our approach models the optically-induced heating of fluid by an illuminated plasmonic nanoantenna. As such, our model comprises of solving the electromagnetic wave equation derived from Maxwell’s equations, the heat transport equation and the Navier-Stokes equation. We simulated the electrothermoplasmonic flow generated via optically-induced heating of a single TiN BNA in the presence of an applied AC electric field. We find that the TiN BNA provides micrometer scale ETP flow velocities with velocities at least three times higher than those of a single Au BNA.

2. Multiphysics modeling

The photothermal heating of the fluid by the illuminated plasmonic nanoantenna results in a temperature gradient in the fluid. This temperature gradient in turn establishes a gradient in the temperature-dependent properties of the fluid namely: the density, permittivity and electrical conductivity. A gradient in the density of the fluid induces buoyancy-driven thermoplasmonic convection. On the other hand, if an AC electric field is applied in the presence of a gradient in the permittivity and electrical conductivity, ETP flow is induced.

The simulation begins by solving the time-independent electromagnetic wave equation given by

$$\nabla \times \nabla \times \mathbf{E} - k_0^2 \varepsilon(\mathbf{r}) \mathbf{E} = 0,$$

where $\mathbf{E}$ is the total electric field in the vicinity of the BNA, $k_0$ is the free-space wavenumber with wavelength $\lambda$, and $\varepsilon(\mathbf{r})$ is the complex, position-dependent ($\mathbf{r}$) permittivity. The solution of Eq. (1) is used to obtain the heat dissipation power density $q(\mathbf{r}) = \text{Re}[\mathbf{J} \cdot \mathbf{E}^*]/2$ [26], where $\mathbf{J}$ is the induced-current density in the BNA and in the underlying metallic film, Re stands for the real part and $\cdot$ is the complex conjugate. The optical properties of TiN and gold were
taken from [47]. The relatively low temperatures considered in this work (below 473 K) are not significant enough to cause an appreciable modulation of the optical properties of TiN and Au as described in [49,50]. Hence, the temperature-independent dielectric properties for TiN and Au at a given wavelength were used in the simulations. The dielectric constant for glass and sapphire substrates were set as 2.10 and 3.06, respectively. The heat source density serves as a source term for the coupled, steady-state heat transfer and fluid mechanics (HT-FM) problem

\[ \nabla \cdot [-\kappa \nabla T(r) + \rho c_p T(r) u(r)] = q(r), \]

\[ \rho (u(r) \cdot \nabla) u(r) + \nabla \rho (r) - \eta \nabla^2 u(r) = F, \]

with \( \nabla \cdot u = 0 \), where \( T(r), u(r) \) and \( \rho(r) \) are the spatial temperature, fluid-velocity and pressure distributions, respectively, and the material parameters are \( \kappa \), \( \rho \), \( c_p \) and \( \eta \) (thermal conductivity, density, heat capacity, and kinematic viscosity, respectively). For TiN, \( \rho = 5400 \text{ kg/m}^3 \), \( c_p = 533.3 \text{ J/(kg·K)} \) and \( \kappa = 60 \text{ W/(m·K)} \), while for Au, \( \rho = 19300 \text{ kg/m}^3 \), \( c_p = 126 \text{ J/(kg·K)} \) and \( \kappa = 0.6 \text{ W/(m·K)} \). For the water medium, \( \rho = 998 \text{ kg/m}^3 \), \( c_p = 4200 \text{ J/(kg·K)} \) and \( \kappa = 0.6 \text{ W/(m·K)} \). The buoyancy-driven natural convection that occurs when only the laser illumination of the BNA is present is described via a volume force given by the Boussinesq approximation,

\[ F_{buoy} = g \rho \beta(T)(T(r) - T_0) \frac{\nabla T}{T}, \]

where \( g \) is the acceleration due to gravity, \( \rho = 998 \text{ kg/m}^3 \) and \( T_0 = 293.15 \text{ K} \) are the reference density of water and temperature, respectively, and \( \beta(T) \) is the temperature-dependent thermal expansion coefficient of water. The assumption in the Boussinesq approximation is that the variations in the density of the fluid have no effect on the flow field, except that they give rise to buoyancy forces. The ETP flow is triggered by applying an AC electric field in addition to the optical illumination. It arises because the AC electric field acts on the heat-induced gradient in the permittivity and conductivity of the fluid element. The additional volume force resulting from the ETP flow \( F_{etp} \) is added to the buoyancy-driven convection volume force density \( F_{buoy} \) given by the Boussinesq approximation described above so that the total force becomes \( F = F_{etp} + F_{buoy} \). The volume force resulting from ETP flow is given by [51]

\[ \left< F_{etp} \right> = \frac{1}{2} \left( \frac{(\alpha - \gamma)}{(1 + \alpha \tau)} \left< \nabla T \right> F_{ac} \right) \left[ \frac{1}{2} \left< (\nabla T)^2 \right> F_{ac}^2 \right], \]

where \( \tau = \varepsilon / \sigma \) is the charge relaxation time of the fluid, which is taken as water, and \( \varepsilon \) and \( \sigma \) are the permittivity and electrical conductivity of the fluid, respectively. The temperature dependence of the permittivity of water is represented as \( \varepsilon(T) = \varepsilon^0 (1 + \alpha (T - T_0)) \), while the temperature dependence of the conductivity of water is represented as \( \sigma(T) = \sigma^0 (1 + \gamma (T - T_0)) \). The electrical permittivity of the water medium at the reference temperature of \( T_0 = 293.15 \text{ K} \) was set as \( \varepsilon^0 = 80 \varepsilon_0 \text{ F/m} \), where \( \varepsilon_0 \) is the permittivity of free space. The electrical conductivity of the water medium at the reference temperature was set as \( \sigma^0 = 50 \mu\text{S/m} \). The expressions \( \alpha = (1/\varepsilon)(\partial \varepsilon / \partial T) \) and \( \gamma = (1/\sigma)(\partial \sigma / \partial T) \) are linear approximations of the temperature dependence of the permittivity and electrical conductivity, respectively and are given as \(-0.004 \text{ K}^{-1} \) and 0.02 \text{ K}^{-1}, respectively [52]. \( E_{ac} \) is the AC electric field in the fluid, which depends on the applied voltage \( v \) across the fluid. The voltage \( v \) used to generate the AC electric field across the water medium varies from 2 V to 6 V, with a frequency of 50 KHz.
The geometry of the simulation domain is depicted in Fig. 1. The electromagnetic problem is solved for a single BNA. The TiN BNA has an altitude of 130 nm, 5 nm tip radius of curvature, a base dimension of 107 nm, and 10.7 nm gap. The TiN BNA is placed on a 120 nm thick TiN film, which is on a glass or sapphire substrate. The top layer above the BNA is immersed in water. The system is illuminated water-side first by a plane wave of a wavelength of 1064 nm with a spot diameter of 1.12 μm. The input plane wave is polarized parallel to the bowtie tip-to-tip axis. The incident power was set to 1 mW, resulting in an incident intensity of $10^7$ W/m$^2$. We make use of a commercial solver (COMSOL Multiphysics) for the simulations. The full-EM domain is a $2.2 \times 2.2 \times 2.2$ μm cube, which includes the physical domain and the PML (perfectly matched layer). The heat transfer and fluid dynamics simulation is solved in a larger $100 \times 100 \times 100$ μm$^3$ domain with a no-slip wall condition, that is $u = 0$ at each boundary [53], and a prescribed temperature of $T_0$. The EM problem uses 4922648 mesh elements and 12.92 GByte RAM, and the HT-FM problem is meshed with an additional 20935490 elements, using 55.83 GByte RAM. The EM problem at a given wavelength took approximately 12 minutes to solve, while the HT-FM problem took 3.5 hours to solve. The simulations were performed on a desktop machine, which has two Intel Xeon processors each with a processor speed of 2.2 GHz and a total of 20 cores. The total RAM is 256 GByte.

3. Results

3.1 Near-field enhancement and light absorption with plasmonic TiN nanoantenna

The 2D electric field spatial distributions around the TiN BNA are depicted in Figs. 2(a) and 2(b) for longitudinal and transverse polarizations, respectively. For longitudinal polarization, most of the electric field is confined near the tips in the gap region of the BNA due to the lightning-rod effect. The enhanced local electric field could be utilized to generate tight trapping potential wells for trapping small nanoparticles. For the transverse polarization, the electric field is no longer concentrated near the gap, but rather the electric field is concentrated on the other vertices of the BNA. Furthermore, the magnitude of the electric field for longitudinal polarization is about an order of magnitude higher than for transverse polarization. The resonance of this BNA system for longitudinal polarization is evident from

![Fig. 1. Geometry of the simulation. A TiN BNA is placed on a TiN film embedded in a thick glass or sapphire substrate and immersed in water. The meshed domains with representative dimensions are shown. A smaller domain was used for the EM simulation, while the heat transfer and fluid dynamics simulations were performed in the larger domain.](image-url)
the spectral electric field enhancement and spectral absorption cross section depicted in Figs. 2(c) and 2(d), respectively, which we have tuned to be near 1064 nm. We also showed the spectral electric field enhancement for an Au BNA whose dimensions have been slightly increased relative to the dimensions of TiN BNA, (while keeping the gap spacing the same) to ensure that it is resonant at the same 1064 nm wavelength. The field enhancement is defined as $|E/E_0|^2$, where $E_0$ is the electric field amplitude of the input plane wave. Figure 2(c) shows the plot of the electric field intensity enhancement as a function of wavelength for optimized BNA designs made of Au and TiN. The electromagnetic field enhancement produced by the Au BNA is higher than that of TiN. However, in terms of the light absorption efficiency, we find that the TiN BNA surpasses the Au BNA as shown in Fig. 2(d). Thus, the TiN BNA will serve as a more efficient nanoscale heat source than the Au BNA.

![Fig. 2. Near field enhancement and absorption cross section spectrum for TiN BNA and Au BNA. The tip-to-tip spacing between the dimer is 10.7 nm, and the thickness is 120 nm. (a) Distribution of the plasmonic hotspot around the TiN BNA on 120 nm thick TiN film on glass substrate with longitudinal polarization. (b) Distribution of the plasmonic hotspot around the TiN BNA with transverse polarization. (c) Local electric field intensity enhancement as a function of wavelength for TiN BNA and Au BNA. (d) Absorption cross section as a function of wavelength for TiN BNA and Au BNA. BNA is bowtie-nanoantenna.](image-url)
3.2 Photothermal response of the plasmonic TiN bowtie nanoantenna

![Image](image-url)

**Fig. 3.** (a) Axial temperature field for TiN BNA on TiN film on a sapphire substrate. (b) Axial temperature field for TiN BNA on TiN film on a glass substrate. The glass substrate enables a better temperature field confinement and a higher temperature rise. The irradiation spot diameter is 1.12 μm. (c) Axial distribution of the temperature rises from the substrate, through the BNA, and into the fluid for TiN BNA on TiN film, and Au BNA on Au film on glass and sapphire substrates. The inset shows the geometry of the system relative to the temperature field. Scale bar is 1000 nm.

We quantified the local temperature rise in the vicinity of the TiN BNA for two different substrates made of glass and sapphire with the polarization of the incident illumination in the longitudinal direction. The spatial temperature field distribution when the TiN BNA on TiN film system on a sapphire substrate was illuminated is depicted in Fig. 3(a). When the substrate was changed to glass, which has a much lower thermal conductivity, the spatial temperature field distribution is depicted in Fig. 3(b). It is evident from Figs. 3(a) and 3(b) that the in-plane confinement of the spatial temperature field is higher for the glass substrate than the sapphire substrate. We attribute this result to the fact that the thermal conductivities of the water medium and glass substrate are much lower than that of TiN so that most of the dissipated heat is confined in the TiN material, with minimal thermal spreading. On the other hand, the thermal conductivity of sapphire is approximately one-half of that of TiN, so that there is more thermal spreading when a sapphire substrate is used. Figure 3(c) is a line plot of the axial (along z-direction) temperature field distribution for TiN BNA on TiN film and Au...
BNA on Au film on glass and sapphire substrates. The maximum temperature rise for all cases occurs on the surface of the BNA. As expected, the TiN BNA on glass substrate generated a high temperature rise of 56 K, while the peak temperature rise on the Au BNA system is only 9 K. This difference is because the TiN BNA on TiN film system absorbs more light than the Au BNA on Au film system. Furthermore, TiN film has a lower thermal conductivity (60 W/mK) than gold (317 W/mK), and hence there is less thermal spreading in the TiN film compared to the Au film. When the substrate was switched to sapphire, the maximum temperature rise on the surface of the TiN BNA reduces to 34 K, while the maximum temperature rise on the Au BNA reduces to 4 K. Furthermore, the use of a high thermal conductivity substrate like sapphire also resulted in a lower temperature rise inside the water medium. Thus, the use of high thermal conductivity substrates represents an elegant approach to manage the temperature rise in integrated nanoplasmonic systems [13]. The use of sapphire as a high thermal conductivity substrate has been recently used to prevent the elevation of the temperature of super-heated water as the optical power of the heating laser is increased [54].

3.3 Enhanced electrothermoplasmonic flow with plasmonic TiN nanoantenna

The heat transferred to the adjoining fluid medium by the hot BNA can generate thermoplasmonic convection flow in the fluid due to the gradient in the density of the fluid. If an AC electric field is applied to the fluid, a much stronger ETP flow is also induced in the fluid. A horizontal 2D slice of the ETP flow velocity profile for the TiN BNA with longitudinal polarization is shown in Fig. 4(a). We see that the flow is radially symmetric. The flow will act to transport suspended objects towards the thermal hotspot, which corresponds to the position of the TiN BNA. A vertical 2D slice of the ETP flow velocity profile for the TiN BNA with longitudinal polarization is shown in Fig. 4(b). In the axial direction, this flow moves in a direction away from the TiN BNA. This implies that in the axial direction, the flow would act to transport suspended objects away from the BNA. Figure 4(c) shows the in-plane radial velocity of the microfluidic flow induced in the fluid at a height of 10 μm from the surface of the BNA. For the case when only the laser illumination of the TiN BNA or Au BNA is applied, the velocity of the induced buoyancy-driven thermoplasmonic convection is extremely weak with a peak of 52.5 nm/s for the TiN BNA and 15.1 nm/s for the Au BNA. Such weak velocities cannot induce a net motion of suspended particles that are constantly undergoing Brownian motion. When an AC voltage of 2 V is applied to induce an electric field within the water medium, the velocity of the resulting ETP flow is enhanced significantly to 0.84 μm/s for TiN BNA and 0.28 μm/s for the Au BNA on glass substrates, respectively. We also find that increasing the voltage by a factor of 3, i.e. from 2 V to 6 V increases the velocity by a factor of approximately 8.5.
Figure 4(d) shows that the ETP flow is also sensitive to the polarization of the incident illumination along the TiN BNA axis. For longitudinal polarization, the maximum radial velocity is 0.84 μm/s, while for transverse polarization, the maximum radial velocity reduces by almost fifty-percent to 0.45 μm/s. This is because, with longitudinal polarization, the BNA absorbs more light and hence induces a larger temperature gradient in the fluid in comparison to when transverse polarization is used.

We also investigated in detail the effect of the substrate on the induced ETP flow. Switching from a glass to a sapphire substrate influences the temperature distribution in the water medium and hence the induced temperature gradient in the fluid. Figure 5(a) shows the in-plane temperature profile in the water medium at a distance of 10 μm from the surface of the TiN BNA. The maximum temperature rise in the water medium when a glass substrate is used is 2.34 K, while the temperature rise is only 0.21 K for a sapphire substrate. Thus, the use of a sapphire substrate reduces the peak temperature rise in the fluid by nearly a factor of 11. This is in correspondence with the change in the maximum velocity of the ETP flow depicted in Fig. 5(c), where it is observed that the maximum ETP flow velocity reduces by a
factor of 11 when the substrate is changed from glass to sapphire. Furthermore, since the volume body force for the ETP flow described in Eq. (4) is proportional to the temperature gradient, it is instructive to check how the ETP flow velocity varies with the temperature gradient in the fluid. Figure 5(b) shows the in-plane temperature gradient along the x-direction in the fluid at a distance of 10 µm from the surface of the TiN BNA. It is evident that the TiN BNA on a glass substrate produces a higher temperature gradient in the fluid in comparison to a sapphire substrate. Thus, the induced ETP flow velocity is expected to be higher when a glass substrate is used as previously shown in Fig. 5(c). We find that changing the substrate from glass to sapphire changes the maximum temperature gradient in the fluid from 0.011 K/µm to 0.094 K/µm, i.e. by a factor of 8.55. This closely agrees with the result in Fig. 5(c), whereby under an applied voltage of 2 V, changing from glass to sapphire reduces the maximum radial velocity from 0.84 µm/s to 0.076 µm/s, which is by a factor of 11. This analysis shows that to enhance the ETP flow, the substrate material should be chosen to maximize the temperature gradient in the fluidic medium.

Fig. 5. (a) Temperature rise in the water medium along the x-direction at a distance of 10 µm from the surface of the TiN BNA. (b) Temperature gradient along the x-direction at a distance of 10 µm from the surface of the TiN BNA. The inset shows the x-direction along which the temperature was obtained relative to the BNA for Figs. 5(a) and 5(b). (c) Maximum radial velocity of the ETP flow under laser illumination and an AC voltage of 2 V and 6 V for TiN BNA on glass and sapphire substrates. (d) Variation of the product of maximum temperature gradient and voltage square with the maximum radial velocity of the ETP flow. A linear variation is obtained both for when the TiN BNA (on TiN film) is on a glass or sapphire substrate.
A closer look at Eq. (4) shows that the volumetric body force for the ETP flow is proportional not only to the temperature gradient, but also to the square of the AC electric field magnitude $|E_{ac}|^2$, and hence it is proportional to the square of the AC voltage $v^2$. Thus, the velocity of the ETP flow will be proportional to the product of the temperature gradient and the square of the applied voltage. Figure 5(d) shows that a plot of the maximum radial velocity versus the product of the maximum of the temperature gradient along the x-direction, and the square of the applied voltage $v^2$ is a straight line for both the glass and sapphire substrates.

4. Conclusion

Using Multiphysics thermodynamics modeling, we have investigated the potential of plasmonic TiN BNA illuminated at near-infrared biological transparency window for plasmon-enhanced optofluidic control in lab-on-a-chip devices. The TiN BNA provides efficient absorption of incident light and generates strong electrothermoplasmonic (ETP) microfluidic flow for on-demand capture and rapid transport of target particles towards the plasmonic hotspots. The TiN BNA generates an ETP flow with a peak velocity that is about three times higher than that of the Au BNA. This makes TiN-based nanoantenna attractive for engineering fluidic flow for directed assembly of analytes, promoting mixing and for enhanced biosensing. We show that for high thermal conductivity substrates where the temperature rise in the fluid is reduced, the ETP flow velocity can be increased by increasing the applied voltage. This follows from the fact that the ETP flow velocity scales as the square of the applied voltage and can be readily amplified by raising the applied voltage. Thus, for temperature sensitive applications, a high thermal conductivity substrate may be used to limit both the temperature rise and the local temperature gradient in the fluid, while the ETP flow is amplified on-demand by raising the applied voltage. Additionally, for applications requiring high optical intensities, the judicious choice of the substrate material can be used to control the thermal heating, while the ETP flow velocity is increased by raising the AC electric field or voltage. Finally, we note that the efficient heat generation enabled by the TiN BNA system also makes it a good candidate material for generating heat-induced forces such as thermophoresis that can be remotely triggered by optical illumination. The theoretical prediction of enhanced ETP flow enabled by plasmonic TiN nanoantennas will be useful for the growing range of lab-on-a-chip devices that utilize plasmonic nanostructures and would open new opportunities for controlling fluid flow on the micro- and nanoscale.

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The authors declare that there are no conflicts of interest related to this article.

References


