Nanoscale Tunable Optical Binding Mediated by Hyperbolic Metamaterials

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Supporting Information

ABSTRACT: Carefully designed nanostructures can inspire a new type of optomechanical interactions and allow surpassing limitations set by classical diffractive optical elements. Apart from strong near-field localization, a nanostructured environment allows controlling scattering channels and might tailor many-body interactions. Here we investigate an effect of optical binding, where several particles demonstrate a collective mechanical behavior of bunching together in a light field. In contrast to classical binding, where separation distances between particles are diffraction limited, an auxiliary hyperbolic metasurface is shown here to break this barrier by introducing several controllable near-field interaction channels. Strong material dispersion of the hyperbolic metamaterial along with high spatial confinement of optical modes, which it supports, allows achieving superior tuning capabilities and efficient control over binding distances on the nanoscale. In addition, a careful choice of the metamaterial slab’s thickness enables decreasing optical binding distances by orders of magnitude compared to free space scenarios due to the multiple reflections of volumetric modes from the substrate. Auxiliary tunable metamaterials, which allow controlling collective optomechanical interactions on the nanoscale, open a venue for new investigations including collective nanofluidic interactions, triggered biochemical reactions, and many others.

KEYWORDS: optical forces, optical tweezers, hyperbolic metamaterials, surface plasmons, optical binding

Optomechanical manipulation1 is a widely used technique across many disciplines,2,3 where it is utilized for many fundamental and applied investigations. The capability to manipulate small objects with focused light beams and measure pico- and even femto-4,5 Newton-scale forces opens a venue for studies of new light–matter interaction regimes6,7 and biomolecular processes8,9 to name just a few. Conventional optical tweezer realizations rely on diffractive optical elements and, as a result, have limited trapping capabilities in application to nanoscale particles. As a promising paradigm solution, auxiliary nanostructures have been introduced. So-called plasmonic tweezers10,11 utilize the capability of noble metal structures to confine light beyond the diffraction limit12,13 and provide improved trap stiffness with relatively low optical powers. While the majority of plasmonic tweezer configurations utilize nanoantenna arrays, optomechanical surfaces,14,15 metasurfaces,16 and metamaterials17,18 have been recently proposed. These types of configurations with less structured features (in comparison to antenna arrays) might provide additional capabilities, such as optical attraction,19 and are less sensitive to accurate positioning of trapping beams with respect to a structure.

In general, functionalities of auxiliary structures can be split into three main categories. The first one is related to the ability of near-field concentration beyond the diffraction limit, which is traced back to the first generation of plasmonic tweezers.20–22 Here the main tool for analysis is based on dipolar approximation, where the manipulated particle’s size is small compared with the fastest spatial intensity variation. It is also important that this model assumes the local field to remain unperturbed by a small particle. The next level of sophistication in auxiliary structure design is to account for a modified density of photonic states, which governs scattering channels from the...
particle. For example, if a nearby structure significantly modifies a scattering pattern, the particle takes the recoil in order to conserve the entire linear momentum. One of the main functions of metasurfaces and metamaterials\textsuperscript{16–18} is to tailor scattering into high density of states modes. Apparently, the most complex approach to optomechanical manipulation utilizes active feedback, where a Brownian particle in an optical field modifies the trapping potential dynamically and experiences a back action effect.\textsuperscript{25}

An important niche in the field of the optomechanical manipulation is devoted to the investigation of many-body interactions mediated by self-consistent optical fields. Light-induced binding of micro- and nanosized objects can provide stable configurations of particles due to light rescattering and their self-organization under external illumination.\textsuperscript{24,25} Capabilities to achieve simultaneous sorting and ordering of particle clusters without a need to structure the incident beam makes optical binding advantageous over holographic tweezing and metamaterial.\textsuperscript{19–21} Various binding scenarios have been investigated and include studies of interactions under Gaussian- and Bessel-shaped beam illumination,\textsuperscript{28,29} pattern creation with several interfering beams,\textsuperscript{30,31} evanescent field excitations, and self-organization of several optically interacting plasmonic particles.\textsuperscript{32} However, those methods rely on either high-field intensities or specific particles’ materials, which may limit their generality. Increasing optical trap stiffness without a need to use high-intensity illumination, flexible control over interparticle distances, and anisotropic optical binding in different directions are among the long-standing challenges, valuable from both fundamental and practical standpoints.\textsuperscript{33} Parameters of optical binding can be significantly influenced by introducing a nearby interface. It modifies both the incident field due to Fresnel reflection and effective particle polarizabilities owing to near-field interactions, qualitatively understood with the help of the image theory.\textsuperscript{20,34,35} It was shown that metal–dielectric interfaces supporting the propagation of surface plasmon-polariton modes (SPPs) can increase optical trapping stiffness and reduce particle–interface separation distances owing to strong interactions with SPPs.\textsuperscript{36–38}

Structured interfaces can provide an additional flexibility in tailoring scattering channels via a predesigned dispersion of surface and bulk modes. Anisotropic response is one among many possibilities. Generally, extremely anisotropic metamaterials\textsuperscript{40–43} have been useful in various types of applications, i.e. cloaking,\textsuperscript{44} super-resolution,\textsuperscript{45,46} and energy transfer,\textsuperscript{47} and recently have opened a venue for flexible optomechanical control. Furthermore, it has been shown that hyperbolic dispersion of bulk modes causes optical pulling forces,\textsuperscript{12} can lead to levitation\textsuperscript{12} and repulsion,\textsuperscript{12} and can even generate negative lateral optical forces along the surface.\textsuperscript{12}

Here we investigate capabilities of hyperbolic metamaterial substrates in application to optical binding. A typical scenario is depicted in Figure 1, where a plane wave illuminates two subwavelength nanoparticles placed in the vicinity of an anisotropic substrate. Particles’ locations in Cartesian coordinates are \((0, 0, a)\) and \((x, y, a)\), where \(a = \lambda_0/30\) is the radius of the particles and \(\lambda_0\) is the incident light wavelength. Light–matter interactions with the particles will be analyzed under the dipolar approximation. There are three types of channels, which govern the binding phenomenon: (i) particle–particle interaction via the substrate modes, (ii) particle–particle interaction via free space modes, and (iii) individual coupling between each particle and the substrate.

Optical force on a particle in the dipolar approximation can be written as follows:

\[
\vec{F} = \frac{1}{2} \mathrm{Re} \left[ \sum_i \left( \alpha(\omega) E(\vec{r}, \omega) \right) \nabla E(\vec{r}, \omega) \right]
\]

where \(E\) corresponds to the \(i\)th component of the self-consistent electric field, \(i = x, y, z\) are coordinates, and \(\alpha(\omega)\) is the dipolar particle’s polarizability in vacuum, including the radiation correction \(\alpha = \frac{a_0}{1 - \frac{a^2}{k^2}}\). \(a_0 = 4\pi\varepsilon_0\alpha^3 \frac{\varepsilon - \varepsilon_z}{\varepsilon + 2\varepsilon_z}\) is the vacuum permittivity, \(\varepsilon_0\) is the permittivity of the particle (we consider \(\varepsilon_0 = 3\)), and \(\varepsilon_z\) is the permittivity of the surrounding medium.

In our notations, where \(\vec{r}_1 = (0, 0, a)\) and \(\vec{r}_2 = (x, y, a)\), the self-consistent electric field is given by

\[
\vec{E}(\vec{r}) = \vec{E}_{inc}(\vec{r}) + \frac{k^2}{\varepsilon_0} G(\vec{r}, \vec{r}_1) \alpha_x \vec{E}(\vec{r}_1)
+ \frac{k^2}{\varepsilon_0} G(\vec{r}, \vec{r}_2) \alpha_z \vec{E}(\vec{r}_2), \quad j = 1, 2
\]

The first term here represents the incident field with the substrate reflection taken into account; the second and the third terms are the contributions of the dipoles. The Green’s function

![Figure 1. General concept of optical binding above a metamaterial slab.](image)

Highly confined optical modes inside the layered hyperbolic metamaterial open additional interaction channels and allow for the formation of dimers and chains with separation distances below the diffraction limit.

This article is organized as follows: Green’s function approach to optomechanical interactions is revised first, followed by the analysis of optical binding next to semi-infinite hyperbolic substrate and finite thickness slab.

### GREEN’S FUNCTION FORMALISM IN APPLICATION TO OPTICAL BINDING NEAR INTERFACES

The considered scenario is depicted in Figure 1, where a plane wave illuminates two subwavelength nanoparticles placed in the vicinity of an anisotropic substrate. Particles’ locations in Cartesian coordinates are \((0, 0, a)\) and \((x, y, a)\), where \(a = \lambda_0/30\) is the radius of the particles and \(\lambda_0\) is the incident light wavelength. Light–matter interactions with the particles will be analyzed under the dipolar approximation. There are three types of channels, which govern the binding phenomenon: (i) particle–particle interaction via the substrate modes, (ii) particle–particle interaction via free space modes, and (iii) individual coupling between each particle and the substrate.
Figure 2. (a) Color map of the imaginary part of the reflection coefficient as a function of the wavevector's x-component and incident wave frequency. From this graph one can, for example, pick out frequencies \( \omega = 2.05 \times 10^{15} \) and \( \omega = 1.4 \times 10^{15} \) rad/s as points A and B, correspondingly (shown with dotted red lines). A corresponds to \( \lambda_0 = 920 \text{ nm} \) and effective medium parameters \( e_\infty = -1.714 + 0.07i, e_\infty = 5.392 + 0.0084i \) and for B \( \lambda_0 = 1350 \text{ nm}, e_\infty = -8.94 + 0.33i, e_\infty = 5.19 + 0.0118i \). (b) Imaginary part of the reflection coefficient as a function of the wavevector x-component. The dependence is plotted for three sets of parameters: A with blue, B with black, and C (ideal case without the SPP contribution for \( \lambda_0 = 920 \text{ nm}, e_\infty = -2 + 0.066i, e_\infty = 0.5 + 0.0084i \)) with red lines. Characteristic regions to underline the contributions of different interaction channels are \( k_x \in (0,k_0) \) for propagating free-space modes, \( k_x \in (k_0,\infty) \) for hyperbolic modes.

Tailoring Green's Functions near Hyperbolic Substrates

Investigation of different particle–substrate interaction channels can be performed by analyzing the corresponding Green's function in reciprocal space (k-space). This integral representation, as it will be shown hereinafter, can be split into three parts corresponding to the interaction channels with different physical origin. In particular, propagating (nonevanescent in the upper half-space) modes, surface plasmons, and bulk hyperbolic modes can be involved. Further, we will consider layered realization of the metamaterial depicted in Figure 1. The permittivity tensor linked to the chosen set of layers is diagonal and obtained via standard homogenization theory with effective medium parameters \( e_\infty = e_z \neq e_x \) where hyperbolic dispersion occurs when \( \text{Re}[e_{xx}] < 0 \) and \( \text{Re}[e_{zz}] > 0 \). Those components also have strong chromatic dispersion, which will be subsequently used for achieving tunability in binding parameters (see section Chromatic Tuning of Binding).

In order to split the spectral integral representing Green's function in k-space, dispersion of the contributing modes should be derived first. The longitudinal component of the wavevector of bulk metamaterial mode has the form

\[
k_{zz} = \sqrt{k_0^2 e_{zz} - k_x^2 e_{xx}}
\]

where \( k_0 \) is the wavenumber of an incident wave and \( k_x \) is the component of the wavevector of a bulk mode along the substrate surface (transversal component). While \( \frac{\text{Re}[e_{xx}]}{\text{Re}[e_{zz}]} < 0 \), the wave propagation in a bulk hyperbolic material is possible as long as \( k_x \) surpasses a critical value

\[
k_{zz} = k_0 \sqrt{e_{zz}}
\]

and \( k_x \) becomes real.

In order to reveal the contribution of different types of modes (free space, plasmons, hyperbolic modes), Fresnel coefficients should be analyzed. The reflection coefficient from a semi-infinite hyperbolic substrate for s- and p-polarized waves is given by

\[
r_p = \frac{e_{xx} k_{zz} - e_{zz} k_x}{e_{xx} k_{zz} + e_{zz} k_x} = \frac{k_{zz} - k_x}{k_{zz} + k_x}
\]

where \( e_{xx} \) denotes the dielectric permittivity of the upper half-space and \( k_{zz}, k_x \) are longitudinal (perpendicular to the substrate) wavevector components in the hyperbolic metamaterial and in the upper half-space, respectively.

Examination of the Fresnel coefficients allows identifying conditions for excitation of two types of modes in the structure: volumetric hyperbolic modes in a metamaterial and surface plasmon-polaritons on its interface. From the reflection coefficient for p-polarization it is possible to obtain the SPP propagation constant (note that SPP is naturally p-polarized):

\[
k_{zz}^{SPP} = k_0 \sqrt{\frac{e_{xx}}{e_{xx} - e_{zz}}}
\]

SPP exists only if \( \text{Re}[e_{xx}] > \text{Re}[e_{zz}] \), resulting in imaginary \( z \) and real \( x \) components of the SPP wavevector. The surface plasmon–polariton resonance condition corresponds to the zero denominator of eq 6, but it is not satisfied as far as \( \text{Re}[e_{xx}] < 0, \text{Re}[e_{zz}] > 0 \). The minimal value of the denominator corresponds to the \( \text{Re}[e_{zz}] \rightarrow 0 \) and \( \text{Re}[e_{zz}] \rightarrow \infty \), which is close to the surface plasmon-polariton excitation. For the opposite case \( \text{Re}[e_{xx}] > 0, \text{Re}[e_{zz}] < 0 \) surface plasmon-polariton does not exist, as perpendicular to the surface wavevector component is real.

Let us consider in detail the hyperbolic case of \( \text{Re}[e_{xx}] < 0, \text{Re}[e_{zz}] > 0 \). Figure 2(a) is presented to provide a better understanding of the modal structure of the system. The imaginary part of the reflection coefficient contains information about all of the modes. Here the dispersion for homogenized multilayered Ag/Ta_2O_5 is presented (filling factor of the structure is 0.133). The imaginary part of the reflection coefficient as a function of the parallel to substrate wavevector component and frequency \( \omega \) is presented. White lines (solid and dash-dotted) correspond to the light line \( (k_0(\omega) = \omega/c) \) and critical wavevector \( k_0c(\omega) \) from eq 4; the dark blue line illustrates the dispersion characteristic of the surface plasmon-polariton \( k_{\text{SPP}}^{\omega} \) from eq 6, and hyperbolic modes are marked with blue
dashed lines (just a few examples). The behavior of the $\text{Im} \left[ r^p \right]$ is in perfect agreement with the dispersion characteristics.

Therefore, there are three important regions governing the interaction of a nanoparticle with the hyperbolic metamaterial. Hyperbolic modes are contributing for $k_x > k_{cr}$ and surface plasmon-polaritons are supported between $k_0$ and $k_{cr}$, so the distance between them defines the overall contribution of SPPs. The free-space modes are allowed at $0 \leq k \leq k_0$. In the particular case of $k_{cr} \rightarrow k_0$, the SPP is negligible while hyperbolic modes and free-space modes play the main role. This scenario (among many others) is considered in Figure 2(b) (red line) to underline the contribution of hyperbolic modes in the absence of SPPs.

Thus, Green’s function for particles–substrate interaction can be decomposed as follows:

$$
\Gamma_{\text{subs}}^{\text{sub}} (\mathbf{r}_1, \mathbf{r}_2) = \int_0^{k_0} \tilde{M}_{\text{subs}}^{\text{sub}} (k_x) dk_x + \int_{k_0}^{k_{cr}} \tilde{M}_{\text{subs}}^{\text{sub}} (k_x) dk_x + \int_{k_{cr}}^{\infty} \tilde{M}_{\text{subs}}^{\text{sub}} (k_x) dk_x
$$

The integrand matrix $\tilde{M}_{\text{subs}}^{\text{sub}}$ in the Green’s function is presented in the Supporting Information (Section Green’s Function Formalism).

In accordance with the aforementioned: $I = \int_0^{k_0}$, the free-space propagating mode contribution, $II = \int_{k_0}^{k_{cr}}$, the surface plasmon-polariton contribution (if SPPs are supported $\varepsilon_{zz} > \varepsilon_i$), and $III = \int_{k_{cr}}^{\infty}$, volumetric (hyperbolic) modes of the substrate.

The hyperbolic mode contribution is usually estimated with the approximation $k_x / k_0 \rightarrow \infty$, where reflection from a substrate depends only on the dielectric permittivities, as long as $k_x > k_{cr.}$ Moreover, the interplay between plasmonic and hyperbolic contributions could be efficiently tailored via adjusting material parameters ($\varepsilon_{zz}$) and, consequently, $k_{cr}$.

In order to demonstrate this capability, the imaginary part of the reflection coefficient for a p-polarized wave as a function of $k_x$ has been plotted in Figure 2(b) for different $\omega$ corresponding to different material parameters: line A ($\lambda_0 = 920 \text{ nm, } \varepsilon_{ss} = -1.714 + 0.075i, \varepsilon_{zz} = 5.392 + 0.0084i$), where $k_{cr}$ is quite large and the SPP contribution is dominating. Line B ($\lambda_0 = 1350 \text{ nm, } \varepsilon_{ss} = -8.94 + 0.33i, \varepsilon_{zz} = 5.10 + 0.0118i$), where SPPs peak is much narrower and bulk mode contribution is more pronounced, and line C for arbitrary metamaterial with ($\lambda_0 = 920 \text{ nm, } \varepsilon_{ss} = -2 + 0.066i, \varepsilon_{zz} = 0.5 + 0.0084i$), where $k_{cr}$ is less than $k_0$, the SPP contribution is absent, and, consequently, the interaction is governed by free space modes and bulk hyperbolic modes (magnitudes are related as $III_2 = 2.5I_1$). These particular scenarios will be further investigated in terms of optical forces. We should stress that each of these integrals is taken into account twice via effective field eq 1, so the overall difference in force is bigger.

Noteworthy, the interval-based integration given by eq 7 is valid only for a standalone particle. Introducing another particle involves cross-coupling between different terms; for example, the SPP generated by the first particle could be scattered by another one into bulk hyperbolic modes and vice versa. This effect will be considered in the next section and shown to have a minor impact on the overall trapping and binding efficiency.

**RESULTS**

Having identified the contribution of different interaction channels to the Green’s function, we can proceed with the self-consistent scattering problem (eq 2).

**Semi-infinite Substrate.** The influence of a semi-infinite anisotropic multilayered metamaterial on optical binding will be analyzed next. The most significant parameters for binding are the period and stiffness, allowing for effective structuring of nanoparticles in many different 2D and even 3D architectures. Recently, we revealed the possibility to bind nanoparticles with subwavelength separation distances via the interference of surface plasmon-polaritons. Introducing additional metamaterial bulk modes seems promising for the further enhancement of binding capabilities.

Let us consider a pair of nanoparticles, one of which is fixed at the origin of the coordinates as in the previous scenario. For the sake of simplicity, we consider the second particle to have the same parameters as the first one. Effective field at the nanoparticle follows from eq 2 and is given in the Supporting Information (Section Expressions for the Effective Fields).

The period of optical binding can now be defined as a distance between two nearest stable equilibrium positions, and the stiffness is the ratio of the restoring force to the particle’s displacement $k = -\Delta F_{\text{p}} / \Delta x$ (in close vicinity to a stable position, where $F_{\text{p}}(x)$ has approximately a linear profile). Hereinafter the period $L_{\text{bind}}$ and distances will be normalized over the incident wavelength $\lambda_0$, and the optical forces are given in pN over intensity (W/μm²).

The material parameters are taken to be the same as for lines A and C in Figure 2(b), corresponding to the dominating

Figure 3. Dependence of the optical binding force on the distance between the particles. (a) is for A from Figure 2(b). (b) is for line C. The blue line corresponds to the total optical binding force near the anisotropic substrate, the red line is for the surface plasmon-polariton mode contribution, the gray circles depict contribution of hyperbolic modes, and the black lines show contribution of free-space propagating modes.
contributions of SPPs (A) and hyperbolic modes (C). In Figure 3 the optical forces for both principally different scenarios are shown. The blue lines correspond to the total optical force, and the black lines correspond to the contribution of modes with \( k_x \leq k_0 \) (propagating free-space modes in the upper half-space). The SPP contribution for A is given by the red line (it is zero for the C case by definition; see the previous section). Hyperbolic mode (HM) contribution is depicted by gray circles.

In case A, the optical forces are fully driven by surface waves, and the contribution of other modes is negligible. In case C despite the predominating influence of the hyperbolic modes, optical binding has almost nothing special in comparison with the free-space scenario. In this case, the HM contribution just increases the force almost twice (which is still two orders of magnitude less than that of SPPs) and slightly shifts the equilibrium positions, almost not affecting \( L_{\text{bind}} \). In this case of a semi-infinite metamaterial for normally incident light the hyperbolic modes excited by the first particle just propagate symmetrically in the volume, not interacting with the second particle, and vice versa.

However, the existing nonzero contribution of the HMs can be explained via the aforementioned cross-terms, when modes excited by one particle are scattered by another one, giving rise to additional HMs with broken symmetry, which, in turn, leads to the optical force shift.\(^{16}\)

Thus, the hyperbolic modes even being dominating in the interaction with the semi-infinite metamaterial do not provide a sufficient contribution to binding in this case.

**Finite Thickness Metamaterial Slabs.** The main reason for the weak influence of HMs on binding is the lack of a feedback from the bulk modes, which propagate away from the particles to infinity. However, as it will be shown hereinafter, strong optical binding can be obtained utilizing an anisotropic finite thickness slab due to reflections of hyperbolic modes from the boundaries. The structure under consideration appears in Figure 4.

![Figure 4](https://example.com/figure4.png)

Figure 4. Scheme of optical binding near an anisotropic hyperbolic metamaterial (HMM) slab. Reflections from the boundaries of the slab form high-intensity regions and result in optical binding with separation distances \( L_{\text{bind}} \) below the diffraction limit.

In contrast to conventional waveguides, \( k_x \) in hyperbolic slabs can achieve rather high values, which, together with the highly confined shape of the modes, could allow for very small distances between the hot-spots driven by multiple reflections. This, in turn, paves the way for strong subwavelength binding of nanoparticles and also provides tunability via changing material parameters, slab thickness, excitation wavelength, etc.

The formalism developed for the semi-infinite substrate is also applicable for the finite-thickness slab. The main difference is in the Fresnel reflection coefficient, which in the latter case is given by

\[
r_{\text{slab}} = \frac{r - r \exp(2ik_xd)}{1 - (r)^2 \exp(2ik_xd)}
\]

where \( d \) represents the thickness of the slab. It is clearly seen that additional periodical maxima will be present in \( r_{\text{slab}}(k_x) \) (reflection of a \( p \)-polarized wave). These peaks are related to the additional boundary, which causes multiple reflections between the upper and lower interfaces. The distance between hot-spots at the interface and, thus, between the bound particles depends on the parameter \( d \) and angle between the group velocity of the hyperbolic modes and the normal to the surface. Noteworthy, hyperbolic modes are not usual “geometric” rays; thus exact calculations are needed to find the actual binding period \( L_{\text{bind}} \).

Let us consider the optical binding force near an anisotropic slab with parameters A and C (from Figure 2(b)) and thicknesses \( d = \lambda_0/2 \) and \( d = \lambda_0/8 \), \( \lambda_0 = 920 \text{ nm} \). Figure 5 represents the imaginary part of the reflection coefficient (left column (a, c)) and optical force (right column (b, d)). Noteworthy, additional peaks corresponding to the multiple reflections appear in the reflection coefficient. Here we show \( \text{Im}[r^2] \) only for \( k_x/k_0 \leq 10 \), because the next peaks are much weaker due to the absorption and are not necessary for subsequent qualitative analysis. However, the force calculations take into account all possible \( k_x (0 \leq k_x/k_0 < \infty) \) to provide accurate values. The distance between the reflection peaks increases (in k-space) with decreasing thickness and leads to the optical force period decrease.

Comparing Figure 2(b) and Figure 5(a) we find SPP contribution to become much less pronounced (magnitudes of integrals from formula (6) \( II \approx 3I \) for a semi-infinite case, \( II \approx 2.3I \) for \( d = \lambda_0/2 \), and \( II \approx 2I \) for \( d = \lambda_0/8 \) (Figure 5(a))) (where I is the integral contribution of the free-space modes from eq 7). For a thin slab \( d = \lambda_0/8 \) the contribution of SPPs \( k_x/k_0 = 1.044 \) and \( k_x/k_0 = 1.84 \) can be considered as negligible for small distances and almost does not influence the optical forces (Figure 5(b)) governed predominantly by the hyperbolic mode with \( k_x/k_0 = 8.8 \) (\( III \approx 7I \)) and \( L_{\text{bind}} \approx 1.8/8 \approx 0.114 \). For \( d = \lambda_0/2 \), however, we have more supported modes, e.g., SPPs \( (k_x/k_0 = 1.17 \text{ and } k_x/k_0 = 1.2) \) together with a set of hyperbolic modes. This leads to the peculiar behavior of the optical forces: HMs with \( k_x/k_0 \geq 3.08 \) enables subwavelength binding with \( L_{\text{bind}} \approx 1/3.08 \approx 0.32 \) modulated by SPPs’ overall envelope (schematically shown with a gray dotted line) with the period \( \sim 0.85 \). Note, all the distances are normalized over the wavelength of the incident wave \( \lambda_0 \).

For the parameters C (Figure 5(c,d)) there is no surface plasmon-polariton contribution by definition (we have chosen the appropriate parameters in the first section especially to emphasize the influence of hyperbolic modes in both semi-infinite and finite cases), because \( k_x/k_0 = 0.7 < 1 \), so the optical forces are completely dependent on the free-space and volumetric modes. For the half-wavelength hyperbolic slab the forces are governed by free-space propagating waves (\( II = 0 \), \( III \approx I \)) with small \( k_x \) and low-amplitude HMs (high \( k_x \) ones are effectively absorbed via a rather large thickness); thus, the dependence is close to optical binding in free space in terms of both period and force magnitude. For the thin slab the force is almost 2 orders of magnitude increased (Figure 5(d)), and the periodicity now is fully driven by the two most pronounced peaks \( k_x/k_0 = 1.33 (L_{\text{bind}} \approx 0.75) \), shown by a gray envelope, and \( k_x/k_0 = 3.11 \), corresponding to \( L_{\text{bind}} \approx 0.32 \) (overall integral \( III \approx 6.3I \)). The total force in both cases reaches 30 fN for intensities of \( \sim 100 \text{ mW/\mu m}^2 \).

Here the stability of equilibrium positions along the x-direction is discussed; however, these positions are stable along...
the y-direction too (see Supporting Information Figure S3). The trapping potential of optical binding approaches $3\kappa T$ at room temperature for both $A$ and $C$ parameters and moderate intensities usually involved in optical trapping of nanoparticles; thus it is stable against Brownian motion. Considering the overall vertical force, we should take into account van der Waals forces attracting the particles to the substrate together with the optical forces. Therefore, the overall force tends to adpress the particles to the surface. However, we should mention here that it can be suppressed or even reversed (if necessary) via, for example, a nanometer-thick dielectric spacer, electrostatic charging, or one more light source, enabling additional optical traps above the surface.

The aforementioned qualitative mode analysis of optical forces and binding period is an approximation. Often, several peaks contribute to the optical force and form a unique signature, either with SPP or not (e.g., curved and asymmetric Figure 5(b,d)). However, it allows for a better understanding of the binding scenarios in the presence of such complicated structures as hyperbolic metamaterials and even for some quantitative estimations provided above. It should be stressed that the existence of higher multipoles in the case of bigger particles leads to more complicated field distribution; therefore, the maxima of the field are less pronounced.

Moreover, comparing two principally different scenarios (with and without SPP contributions), we find a new possibility to obtain optical binding forces, which are several orders of magnitude higher than in the free-space binding scenarios (and about one order of magnitude stronger than that delivered by plasmonic metals). Furthermore, tuning the distance between

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**Figure 5.** Imaginary part of the reflection coefficient over relative transversal wavevector component (a, c) and optical binding force (b, d). The dependence is plotted for two sets of parameters: (a) and (b) correspond to the parameters $A$ from Figure 2. (c) and (d) correspond to $C$ from Figure 2. The solid lines depict the reflection coefficient and optical binding force for a slab with thickness $d = \lambda_\text{0}/2$; the dash-dotted lines depict the reflection coefficient for a slab with the thickness $d = \lambda_\text{0}/8$, where $\lambda_\text{0} = 920$ nm is the incident wavelength. The black arrows in (a) and (c) show the position of the $k_\text{cr}$. One can see that for $C$ it is placed before $k_\text{0}$. Gray dotted lines represent envelopes (see text). The insets in (b) and (d) show electric component $E_x$ of the field scattered by the particle above the homogeneous hyperbolic substrate with thickness $d = \lambda_\text{0}/2$ (left column) and $d = \lambda_\text{0}/8$ (right column).

**Figure 6.** Chromatic tuning for the multilayered structure of silver and Ta$_2$O$_5$ layers with a slab thickness 115 nm and filling factor of 0.133. (a) Imaginary part of the reflection coefficient (dispersion diagram). (b) Optical binding period over the frequency.
bound particles beyond the diffraction limit is also possible. These characteristics are strongly enhanced in the case of thin slabs, allowing for better utilizing hyperbolic mode feedback, and paves the way to a plethora of highly demanded applications.

Chromatic Tuning of Binding. In the previous sections we have considered semi-infinite and finite slabs of hyperbolic metamaterials consisting of Ag$^{59}$ and Ta$_2$O$_5$ layers, allowing for effective tuning via adjusting material and geometrical parameters. Hereinafter let us consider another important degree of freedom: chromatic tuning of the metamaterial-assisted optical binding.

Figure 6(a) shows the dependence of the reflection coefficient on the incident wave frequency and $k_0/k_f$ for a slab thickness of $115$ nm. It can be seen that the number of HM peaks governed by the reflections (equal to Fabry–Perot resonances for hyperbolic modes) and contributing to optical binding is increased with lower frequency ($d/\lambda_0$ decrease), so the optical force dependence becomes more complicated.

The distance between the bound particles (Figure 6(b)) is now a function of the frequency; thus the material dispersion plays a key role here. The binding period is proportional to the thickness of the incident slab and incident wavelength and $\varepsilon_{22}(\omega)$ and $\varepsilon_{11}(\omega)$. In this case the permittivities are monotonically dependent on the frequency; thus the dependence of the optical binding distance is more or less monotonic. However, in other wavelength regions additional HMs and nonmonotonic dispersion of optical constants could displace the stable equilibrium positions and change the dependence shown in Figure 6(b). This additional degree of freedom opens room for opportunities for tuning optical binding via a “noninvasive” way and fabricate novel designs and architectures of nanostructures on metamaterial substrates by adjusting optically induced forces with hyperbolic modes.

Moreover, in the Supporting Information (Section The Impact of the Topmost Layer) we consider the dependence of the optical binding force on the topmost layer of the slab (metal or dielectric), which also can be useful in a plethora of applications.

CONCLUSION

In this work we describe the transverse optical binding of two particles near a hyperbolic metamaterial. High-$k$ volumetric modes can provide additional channels of the particles’ interaction with substrates and, therefore, drastically enhance capabilities of optomechanical manipulation schemes. For a semi-infinite (or rather thick) metamaterial slab the hyperbolic modes even being dominating in scattering do not contribute to optical binding because of the almost absent feedback (hyperbolic modes excited by one particle do not interact with the second one). In contrast, thin metamaterial slabs provide multiple reflections from boundaries, forming a set of strongly localized hot-spots with huge intensity gradients governing nanoparticle motion at the nanoscale. Furthermore, mode analysis shows the predominant impact of HMs on binding giving rise to several orders of magnitude increased optical forces and deeply subwavelength nanoparticle positioning. Moreover, the principal realization of this phenomenon appears to be rather tolerant to the metamaterial parameters, enabling strongly enhanced performance for a whole set of designs and driving broadband chromatic tuning. Novel auxiliary carefully designed metamaterials and metasurfaces featuring superior optomechanical mechanisms are nowadays extremely demanded in a variety of applications, such as microfluidics, lab-on-a-chip devices, and biology and medicine to name just a few.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsphotonics.9b01378.

1. Green’s function formalism; 2. Expressions for the effective fields; 3. The impact of the topmost layer; 4. Spatial distribution of the electromagnetic field above the thin slab; 5. The stability along the $y$ directions (PDF)

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N.K. designed the model and the computational framework and analyzed the data. A.P. and D.K. performed finite element simulations. A.S.S. and P.G. wrote the manuscript and supervised the work. All authors contributed to the research activities.

Notes

The authors declare no competing financial interest.

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