# nature portfolio

# Peer Review File

# Morphology-independent general-purpose optical surface tractor beam



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#### **REVIEWER COMMENTS**

#### **Reviewer #1 (Remarks to the Author):**

In this manuscript, the authors offer a thorough analysis regarding the optical force generated by a surface wave (SW). Their theory serves as the foundation for the development of a universal optical surface tractor beam (OSTB) capable of attracting any passive particle, irrespective of its characteristics such as size, composition, or geometry. The idea of practical free space optical tractor beams has been introduced more than one decade ago and its experimental verification was demonstrated in 2013. However, passive tractor beams work for very short distances to pull the very small size particles. Later, active tractor beams were introduced for practical applications. In last ten years, the literature of tractor beams has experienced a lot of works including the pulling of cluster or mixture of small sized particles using surface waves. The idea of surface wave tractor beams has also been introduced long ago. However, the main claim of this article:

Instead of increasing the momentum of the transmitted/scattered light beam from a particle (in comparison with the incident beam), the momentum of incident light beam (which is transferred to the scatterer) has been designed negative (artificially with DNG Metamaterial) to pull the scatterer. This is an interesting addition in the literature of tractor beams. Authors have presented a detailed mathematical analysis.

However, I have found several problems regarding the demonstration of their claim. I was searching the practical design of the surface which creates surface wave that transfers negative optical momentum to the scatterer. I did not find the practically proposed design of metamaterial surface in their main article. Later, their proposed possible realistic design has been found in the figures of the supplement article. It seems that such a metamaterial is not practical enough to pull a particle over its surface. If such holes take place over a surface, how the scatterer will be pulled (even a short distance)? Previous surface wave tractor beams pulled scatterers (not only a single particle; all types of particles including cluster of particles) over planar surfaces. If metamaterials were used, previous authors tried to pull the particles inside the metamaterial [Shalin, Alexander S. et al. "Optical pulling forces in hyperbolic metamaterials." Physical Review A 91, no. 6 (2015): 063830] or gain

medium ["Negative electromagnetic plane-wave force in gain media." Physical Review E 84, no. 5 (2011): 057602.] or inside phonic crystal structures. Such previously reported designs (even though theoretical works) seem OK because those designs did not use 'metamaterial surface' to pull the scatterers. Notably, the surface of specially designed hyperbolic metamaterial has been used previously to pull a scatterer over a planar surface and it is quite practical to observe the tractor beam effect over such a planar hyperbolic metamaterial surface [Jin, Renchao et al. "Optical pulling forces enabled by hyperbolic metamaterials." Nano Letters 21, no. 24 (2021): 10431-10437]. Though I appreciate the mathematical analysis of the authors (this article) and the main idea (transfer of negative incident momentum to the scatterer) of this article, I have serious doubt about the practical implementation. If this work 'were' an experimental work, I would recommend for publication. But I have serious doubt about the overall mechanism (I will explain next) and its practical application/possibility of implementation in future.

Most important drawbacks:

Authors have used the word 'canonical momentum' instead of 'Minkowski momentum' to describe the 'wave momentum'. I probably understand the reason. The 'transferred' momentum of light inside a metamaterial or over its surface is still a controversial topic [ See: Kemp, Brandon A. et al. "Electromagnetic and material contributions to stress, energy, and momentum in metamaterials." Advanced Electromagnetics 6, no. 1 (2017): 11-19]. Considering the aforementioned fact, I have huge doubt regarding the final full wave simulations of the article. What I have understood: authors have not even incorporated the actual metamaterial structure (which they have shown in the supplement article) to demonstrate the optical pulling effect. I have felt that the authors have put only the necessary parameters of a possible/ theoretical metamaterial to a hypothetical 3D rectangular structure (their figure-1) in COMSOL and using that structure, all the full wave simulations have been demonstrated. Unfortunately, this may not be good enough to support such a remarkable claim of transferring negative canonical momentum to the scatterer and universal OSTB. I humbly request the authors to design (for full wave simulation in COMSOL) a practically realizable metamaterial surface, which will really support their 'big' claim. Even if the authors would utilize the structures (given in their

supplement article) for full wave simulations to demonstrate the optical pulling force, it would be an important support for their claim. However, this is not the case (they have used planar surface). Even the recent metasurafce or hyperbolic metamaterial or other surface wave-based works on optical force consider the full wave simulations of particles putting them over the realistic design [for example- (1) Petrov, Mihail I et al ."Surface plasmon polariton assisted optical pulling force." Laser & Photonics Reviews 10, no. 1 (2016): 116- 122. Or (2) Li, Tianyue et al."Reversible lateral optical force on phase-gradient metasurfaces for full control of metavehicles." Optics Letters 48, no. 2 (2023): 255-258] or (3) Jin, Renchao et al. "Optical pulling forces enabled by hyperbolic metamaterials." Nano Letters 21, no. 24 (2021): 10431-10437]. Such realistic simulation results/verifications are fully missing in this work (even though the main idea is interesting). My suggestion: Figure-1 should include at least two more figures describing the real design of the metamaterial surface (showing where the actual particle(s) is placed) along with the clear identification of the incident angle of light beam (also the polarization). This paper mentions the development of a more advanced optical surface tractor beam, but it lacks specific realistic setup details.

I believe there are some other issues with this manuscript:

1. The Authors claim optical surface tractor beam (OSTB) that can pull virtually any passive particle, regardless of its size, composition, and geometry. However, it is noteworthy that the manuscript lacks any numerical simulations or discussions specifically addressing particle size (Mie or Rayleigh or Dipolar ??) putting them over a realistic metamaterial surface.

2. The manuscript does not include a discussion of chiral or plasmonic particles or whether the optical surface tractor beam (OSTB) can effectively interact with diverse particle types, such as lossy, lossless, or chiral, when they coexist within a cluster or mixture.

3. Very important: The authors say that COMSOL simulations are converged, but in numerical works some details are always necessary, to allow others to reproduce the results. Details about the mesh and solvers etc. can be added in the Supporting Information. 4. Most important one: The authors should address a discussion regarding the incident angle of light in their manuscript. What is the laser beam's incident angle? If anyone alters incident angle or modify the propagation and polarization, would the results remain consistent?

5. The introduction section does not clearly state where and how this technology can be applied and the potential benefits it offers.

I believe that focusing on these areas would definitely improve the manuscript. Most importantly, this article should be re-written properly so that it can be understood in a much better way. Some notable parts of the supplement (realistic design and force on the scatter while placing over such a realistic metamaterial) should be put in the main article. As this is not an experimental work, at least full wave simulation results of optical force on a (or multiple) scatterer over a realistic or more practical real metamaterial surface is a necessity.

- Considering all the aforementioned issues; at this moment, I am failing to recommend this version of the article for publication in Nature Communications.

#### **Reviewer #2 (Remarks to the Author):**

This manuscript proposes a new mechanism to achieve optical pulling force by using a special type of surface wave with negative wavevector (phase velocity). This surface tractor wave can be realized by engineering the material parameters (permittivity and permeability) of the substrate so that they satisfy certain condition, corresponding to equation (18) or (20). Under this condition, the incident surface wave carries negative canonical momentum with a single well-defined negative k. The authors rigorously proved, via analytical theories based on Minkowski stress tensor and numerical simulations, that the surface tractor beam can pull any particle on the surface towards the source. Remarkably, this negative force works for particles of any size and geometry, and it can be enhanced by absorption (which typically destroys the pulling force in conventional systems). Realistic design of the substrate with the required material parameters has been provided, and the motion of a particle under the pulling force as well as its trajectory have also been discussed.

I find the mechanism of realizing a pulling force reported in this work novel, because it is robust to material absorption and works on various types of particles, which cannot be achieved with previously reported mechanisms strongly relying on engineering the scattering channels. This is an essential step towards practical applications of optical pulling force. The authors carefully addressed the issue of Abraham-Minkowski momentum debate, making the analysis in the work rigorous. I believe the results will be of broad interest to the optics community and others. The manuscript is well prepared and clearly structured. I am pleased to recommend it for publication in Nature Communications, and I have the following minor comments for the authors' consideration.

1. The authors have studied the pulling force on dielectric particles in detail. Does the mechanism apply to metallic particles of arbitrary size?

2. It seems to me that the result of Eq. (15) relies on the result of Eq. (17). Will it be better to re-arrange the derivations for the two equations?

3. The lateral optical forces also strongly depend on the surface waves or guided waves of the substrate. Can the mechanism be applied to reverse the lateral optical force?

4. In experiments, there are usually many particles on the substrate. Will the pulling force be affected by the multiple scattering of the particles?

#### **Response to Reviewers**

Reviewer #1 (Remarks to the Author):

In this manuscript, the authors offer a thorough analysis regarding the optical force generated by a surface wave (SW). Their theory serves as the foundation for the development of a universal optical surface tractor beam (OSTB) capable of attracting any passive particle, irrespective of its characteristics such as size, composition, or geometry. The idea of practical free space optical tractor beams has been introduced more than one decade ago and its experimental verification was demonstrated in 2013. However, passive tractor beams work for very short distances to pull the very small size particles. Later, active tractor beams were introduced for practical applications. In last ten years, the literature of tractor beams has experienced a lot of works including the pulling of cluster or mixture of small sized particles using surface waves. The idea of surface wave tractor beams has also been introduced long ago. However, the main claim of this article:

Instead of increasing the momentum of the transmitted/scattered light beam from a particle (in comparison with the incident beam), the momentum of incident light beam (which is transferred to the scatterer) has been designed negative (artificially with DNG Metamaterial) to pull the scatterer. This is an interesting addition in the literature of tractor beams. Authors have presented a detailed mathematical analysis.

We thank Reviewer #1 for accurately summarizing our work. The concerns of the reviewers are addressed in a point-by-point manner below.

However, I have found several problems regarding the demonstration of their claim. I was searching the practical design of the surface which creates surface wave that transfers negative optical momentum to the scatterer. I did not find the practically proposed design of metamaterial surface in their main article. Later, their proposed possible realistic design has been found in the figures of the supplement article. It seems that such a metamaterial is not practical enough to pull a particle over its surface.

We thank Reviewer #1 for raising this very important issue of designing a realistic and practical metamaterial for pulling. As the Reviewer has pointed out, the metamaterial is characterized by effective homogeneous permittivity and permeability in our initial simulations. It would be certainly better if the pulling simulation incorporates the actual nanostructures that constitute the metamaterial.

We have added such realistic simulations to the manuscript. One can see that TM-polarized OSTBs capable of pulling particles can be generated on our realistic metamaterial illustrated in the inset of Fig. 4a by using either the line sources approach (Fig. 5a) or the prism approach (Fig. 5d). The possession of negative wavevector by the surface wave such-generated is clearly illustrated in Fig. 5c. Then, in full-wave simulations that fully consider the nanostructure, such surface wave is demonstrated to be capable of pulling particles of various shapes (circular, square, and triangular) and compositions (dielectric, chiral, metallic, and absorptive), as shown in Fig. 5e-g. We stress that, unlike our previous pulling force calculations that were done with a surface characterized by a homogeneous permittivity and permeability, these pulling forces are calculated for a single or a cluster of particles on top of a realistic nanostructure, as suggested by the Reviewer.

The newly added Fig. 4 is shown below, which shows the actual unit cell of our nanostructure (inset of a), as well as its electromagnetic properties. The red circle line in Fig. 4c displays the dispersion of the surface state supported in the air/metamaterial interface. It's evident that this surface state exhibits a negative group velocity for a positive wavenumber, precisely corresponding to the OSTB we proposed.



Fig. 4. **a** The dispersion relation for the TM modes. The unit cell of the metamaterial is shown in the inset, with cyan regions and the white circles representing the background medium (*εh*=120) and air holes (radius of  $r_c$ =0.1*a*), respectively. **b** Effective constitutive parameters for the red-highlighted band segment in panel **a**. **c** The projection of the bulk bands (gray disks) and the surface band (red circles). The normalized electric field in a supercell is shown in the rectangular bar on the right, where the black disks and the dashed line represent the air holes and the photonic crystal/air interface, respectively.

The newly added Fig. 5 illustrates (a-d) how the surface wave can be generated either by a series of parallel line sources or by illuminating a prism, and (e-f a) how the surface wave pulls various particles.



Fig. 5. **a** A TM OSTB is excited above a photonic crystal (consisting of a squared array of air holes represented by black disks and with a period of 100 nm) by using a series of line sources (each with an amplitude of 1 mA and represented by a white star). The vacuum wavenumber is  $k_0 = 0.527/a$ . The line sources are located within the rectangle formed by the black dashed lines. A zoomed-in view is shown in **b**. **c** The Fourier transform spectra of the electric fields on the metamaterial surfaces along the green dashed lines in panels **a** and **d**, indicating the successful generation of a surface wave with a negative wavevector. **d** A TM OSTB propagating along the surface of the metamaterial consisted of realistic microstructures excited by illuminating a prism (outlined by the dotted lines, refractive index=1.69) at an incident angle of 45°. **e** The longitudinal optical forces acting on circular particles of different sizes as functions of δ, where ε*r*=5 and *h'=a*. **f** The longitudinal optical forces of the dielectric ( $\varepsilon_r$ =5), plasmonic ( $\varepsilon_r$ =-3+0.5*i*), and chiral ( $\varepsilon_r$ =5,  $\chi$ =1.0) circular particles versus their dimensionless particle size *k*0*r*. The forces of plasmonic particles are multiplied by 10 for clarity. **g** Pulling forces on circular (transparent), square (lossy) and triangular (chiral) shaped particles arranged equidistantly, where  $k_0r = 1.5$ , Re( $\varepsilon_r$ )=9.

If such holes take place over a surface, how the scatterer will be pulled (even a short distance)? Previous surface wave tractor beams pulled scatterers (not only a single particle; all types of particles including cluster of particles) over planar surfaces. If metamaterials were used, previous authors tried to pull the particles inside the metamaterial [Shalin, Alexander S. et al. "Optical pulling forces in hyperbolic metamaterials." Physical Review A 91, no. 6 (2015): 063830] or gain medium ["Negative electromagnetic plane-wave force in gain media." Physical Review E 84, no. 5 (201Figf): 057602.] or inside phonic crystal structures. Such previously reported designs (even though theoretical works) seem OK because those designs did not use 'metamaterial surface' to pull the scatterers. Notably, the surface of specially designed hyperbolic metamaterial has been used previously to pull a scatterer over a planar surface and it is quite practical to observe the tractor beam effect

over such a planar hyperbolic metamaterial surface [Jin, Renchao et al. "Optical pulling forces enabled by hyperbolic metamaterials." Nano Letters 21, no. 24 (2021): 10431- 10437].

We thank Reviewer #1 for raising this issue. In fact, the air holes are aligned along the *y*-axis, while the air/metamaterial interface is on the xy plane, which is flat in our practical design, as shown in the insets of Fig. 5**e** and 5**g**. The interface is flat and mechanically stable to the particle, and the particle being pulled will neither be impeded nor trapped by the air holes, see Fig. 5e. If we wish, we can fill the air hole with low dielectrics. As depicted in Fig. R1, when we adjust the radius of the air holes and fill them with low dielectrics, the OSTB (red circle line in Fig. R1b) can also be supported in the air/metamaterial surface.

To emphasize the importance of the issue, we have cited the above-listed papers in our manuscript.



Fig. R1. **a** The band dispersion of a photonic crystal composed of glass cylinders embedded inside the dielectric matrix. **b** The projection of bulk bands (gray disks) and the surface band (red circles).

Though I appreciate the mathematical analysis of the authors (this article) and the main idea (transfer of negative incident momentum to the scatterer) of this article, I have serious doubt about the practical implementation. If this work 'were' an experimental work, I would recommend for publication. But I have serious doubt about the overall mechanism (I will explain next) and its practical application/possibility of implementation in future.

We thank Reviewer #1 for appreciating our mathematical analysis. We believe that our mathematical analysis firmly established the working principles for our surface wave pulling. The reviewer has raised concerns about the overall mechanism, particularly due to its practical application/possibility of implementation in the future. We hope that Reviewer #1 will appreciate the practicality of our design after reading this reply.

Most important drawbacks:

Authors have used the word 'canonical momentum' instead of 'Minkowski momentum' to describe the 'wave momentum'. I probably understand the reason. The 'transferred' momentum of light inside a metamaterial or over its surface is still a controversial topic [ See: Kemp, Brandon A. et al. "Electromagnetic and material contributions to stress, energy, and momentum in metamaterials." Advanced Electromagnetics 6, no. 1 (2017): 11- 19].

We agree with the Reviewer that the momentum of light inside a metamaterial or over its surface is controversial. Nevertheless, as Reviewer #1 has probably noted, such controversy did not affect our discussion or conclusion. Since the particle is always immersed in air, our derivation starts from a rigorous calculation of optical force and introduces no unjustified approximations. In fact, our work partly resolved the controversy by proving that the 'momentum' responsible for the longitudinal force is precisely the Minkowski or canonical momentum. According to the definitions of canonical momentum density and group velocity, the expression of longitudinal force (Eq. (6)) can be transformed into (see Supplementary Equations (S42) and (S43))

$$
F_x = -\iint\limits_{S_x} v_g^s g_x^{is} dS - \iint\limits_{S_x} v_g^s g_x^s dS - \iint\limits_{S_x} v_g^f g_x^f dS, (R1)
$$

where  $v_g^s$ ,  $v_g^f$  are the group speeds of the surface wave and freely propagating waves, and

 $g_x^i$ ,  $g_x^s$ ,  $g_y^f$  are the canonical momentum densities of the incident surface wave that is being

intercepted (scattered or absorbed), scattered surface wave, and freely propagating waves, respectively. The first term in Eq. (R1) denotes the canonical momentum flux of intercepted incident photons, while the second and third terms represent the canonical momentum flux of reemitted photons. Eq. (R1) reveals that the longitudinal optical force induced by the surface wave is attributed to the transfer of canonical momentum.

#### We have cited the listed papers as references for the controversy in electromagnetic momentum.

Considering the aforementioned fact, I have huge doubt regarding the final full wave simulations of the article. What I have understood: authors have not even incorporated the actual metamaterial structure (which they have shown in the supplement article) to demonstrate the optical pulling effect. I have felt that the authors have put only the necessary parameters of a possible/ theoretical metamaterial to a hypothetical 3D rectangular structure (their figure-1) in COMSOL and using that structure, all the full wave simulations have been demonstrated. Unfortunately, this may not be good enough to support such a remarkable claim of transferring negative canonical momentum to the scatterer and universal OSTB. I humbly request the authors to design (for full wave simulation in COMSOL) a practically realizable metamaterial surface, which will really support their 'big' claim. Even if the authors would utilize the structures (given in their supplement article) for full wave simulations to demonstrate the optical pulling force, it would be an important support for their claim. However, this is not the case (they have used planar surface). Even the recent metasurafce or hyperbolic metamaterial or other surface wave-based works on optical force consider the full wave simulations of particles putting them over the realistic design [for example- (1) Petrov, Mihail I et al ."Surface plasmon polariton assisted optical pulling force." Laser & Photonics Reviews 10, no. 1 (2016): 116-122. Or (2) Li, Tianyue et al."Reversible lateral optical force on phase-gradient metasurfaces for full control of metavehicles." Optics Letters 48, no. 2 (2023): 255-258] or (3) Jin, Renchao et al. "Optical pulling forces enabled by hyperbolic metamaterials." Nano Letters 21, no. 24 (2021): 10431-10437]. Such realistic simulation results/verifications are fully missing in this work (even though the main idea is interesting).

Indeed, what the Reviewer described is exactly what we were doing. We have followed the suggestion of the Reviewer to perform full-wave simulations with the particle being pulled on top of a photonic crystal nanostructure, which put our former conclusions on much firmer ground. We believe such simulations with realistic nanostructures have strengthened the credibility and practicality of our work, as suggested by the reviewer. We have now cited the listed references in the manuscript to illustrate the importance of realistic modeling of the metamaterial.

My suggestion: Figure-1 should include at least two more figures describing the real design of the metamaterial surface (showing where the actual particle(s) is placed) along with the clear identification of the incident angle of light beam (also the polarization). This paper mentions the development of a more advanced optical surface tractor beam, but it lacks specific realistic setup details.

We fully agree with Reviewer #1's suggestion. Considering the overall structure of the manuscript, we prefer to first lay down the analytic theory before discussing its practical implementation with realistic metamaterials. As such, the figures requested by Reviewer #1 are inserted as Fig. 4 and Fig. 5 in the manuscript. The optimal incident angle for light beam, which is chosen to phase match the surface wave is illustrated in Fig. 5d for the case of surface wave excitation by a prism.

I believe there are some other issues with this manuscript:

1. The Authors claim optical surface tractor beam (OSTB) that can pull virtually any passive particle, regardless of its size, composition, and geometry. However, it is noteworthy that the manuscript lacks any numerical simulations or discussions specifically addressing particle size (Mie or Rayleigh or Dipolar ??) putting them over a realistic metamaterial surface.

We thank Reviewer #1 for pointing this out. We have now included realistic simulations that consider the microstructure of the metamaterial for particles of different shapes (circular, triangular, square, or cluster, see Fig. 5g), composition (dielectrics, plasmonics, and chiral, see Fig. 5f) and sizes (dipolar to Mie regime, see Fig. 5f) in the manuscript. Our new simulations provide strong evidence supporting the conclusions of our analytical theory, namely that the OSTB pulls all kinds of passive particles.

2. The manuscript does not include a discussion of chiral or plasmonic particles or whether the optical surface tractor beam (OSTB) can effectively interact with diverse particle types, such as lossy, lossless, or chiral, when they coexist within a cluster or mixture.

We thank the Reviewer for his/her valuable suggestion. We have now performed simulations to further support our conclusion. For particle composition, we have checked chiral, plasmonics, dielectric, and lossy particles (Fig. 5f). For particle shape, we have checked circular, triangular, or square particles (see Fig. 5g). For particle size, we have checked particle with size ranging from dipolar to Mie particles (see Fig. 5f). We have also checked mixtures of lossless, lossy, and chiral particles (see Fig. 5g and Supplementary Figure 6). In all cases, pulling forces are observed, just as expected from our analytical theory.

3. Very important: The authors say that COMSOL simulations are converged, but in numerical works some details are always necessary, to allow others to reproduce the results. Details about the mesh and solvers etc. can be added in the Supporting Information.

We certainly agree with the Reviewer. We have now provided the simulation details in the Method section in the manuscript.

4. Most important one: The authors should address a discussion regarding the incident angle of light in their manuscript. What is the laser beam's incident angle? If anyone alters incident angle or modify the propagation and polarization, would the results remain consistent?

We have performed simulations for generating a surface wave using line sources or a prism. In the particular case of a prism shown in Fig. 5d, the optimal angle for excitation is 45<sup>o</sup> (depending on the shape and refractive index of the prism), where the wavevectors of the incident wave and the surface wave match. In our design, 45 degrees is ideal for excitation, see the newly added Supplementary Figure 4. When the incident angle deviates from 45 degrees, or the incident beam is not perfectly TM-polarized, the excitation efficiency will be reduced slightly, as evident by the Fourier spectra of the excited surface modes in panels **c** and **d**. As long as the deviation of the incident angle from 45 degrees is not significant, the surface wave can still be generated, but with a somewhat reduced amplitude. In the supplementary information, the discussion about the incident angle and polarization of light is added.



**Supplementary Figure 4.** TM OSTBs are excited using prisms. **a** and **b,** The y-component electric field distributions. The black dashed line outlines the prism boundary, while white arrows indicate the directions of incident Gaussian beams. Both prisms have a refractive index of 1.69. **c** and **d,** The Fourier transform spectra of the electric fields on the metamaterial surface  $(70a < x < 110a, z = 16a)$ . In panel **c**, the black and red solid lines correspond to perfect TM-polarized and hybrid (81% TM + 19% TE) polarized incident beams, respectively. All incident beams have the same amplitude.

5. The introduction section does not clearly state where and how this technology can be applied and the potential benefits it offers.

We have followed the Reviewer's suggestion by explicitly verifying that our universal surface tractor beam works for a realistic metasurface. Given the robustness of our theory (works for a single particle or cluster of particles, irrespective of size and composition, etc.), the experimental implementation will not require delicate adjustment. This is in sharp contrast with previous works on tractor beams, where careful design of the particle and beam is necessary. Thus, our OSTB is more practical than the previous tractor beam.

The fact that our OSTB works near a surface makes it compatible with lab-on-a-chip optofluidic devices, where one could simultaneously pull a collection of particles backward without having to worry about particle morphologies or distribution.

In the revised manuscript, we have added a short paragraph at the end of the introduction section to discuss the application of our technology:

"The proposed OSTB, owing to its ability to pull arbitrary particle or particle clusters (where the entire cluster can be viewed as a single particle), can be applied to manipulate particles in an optofluidic or lab-on-a-chip environment, where one could simultaneously pull a collection of

#### particles backward without having to worry about particle morphologies or distribution."

I believe that focusing on these areas would definitely improve the manuscript. Most importantly, this article should be re-written properly so that it can be understood in a much better way. Some notable parts of the supplement (realistic design and force on the scatter while placing over such a realistic metamaterial) should be put in the main article. As this is not an experimental work, at least full wave simulation results of optical force on a (or multiple) scatterer over a realistic or more practical real metamaterial surface is a necessity. I am running a few minutes late; my previous meeting is running over.

We followed the reviewer and performed a full-wave Comsol simulation on realistic metamaterials. We have restructured the manuscript for clarity and moved the part on realistic design to the main text. We thank the reviewer for his/her constructive suggestions.

- Considering all the aforementioned issues; at this moment, I am failing to recommend this version of the article for publication in Nature Communications.

We hope that our effort to address all the Reviewers' comments will make Reviewer #1 consider our manuscript good enough for publication in Nature Communications.

#### Reviewer #2 (Remarks to the Author):

This manuscript proposes a new mechanism to achieve optical pulling force by using a special type of surface wave with negative wavevector (phase velocity). This surface tractor wave can be realized by engineering the material parameters (permittivity and permeability) of the substrate so that they satisfy certain condition, corresponding to equation (18) or (20). Under this condition, the incident surface wave carries negative canonical momentum with a single well-defined negative k. The authors rigorously proved, via analytical theories based on Minkowski stress tensor and numerical simulations, that the surface tractor beam can pull any particle on the surface towards the source. Remarkably, this negative force works for particles of any size and geometry, and it can be enhanced by absorption (which typically destroys the pulling force in conventional systems). Realistic design of the substrate with the required material parameters has been provided, and the motion of a particle under the pulling force as well as its trajectory have also been discussed.

I find the mechanism of realizing a pulling force reported in this work novel, because it is robust to material absorption and works on various types of particles, which cannot be achieved with previously reported mechanisms strongly relying on engineering the scattering channels. This is an essential step towards practical applications of optical pulling force. The authors carefully addressed the issue of Abraham-Minkowski momentum debate, making the analysis in the work rigorous. I believe the results will be of broad interest to the optics community and others. The manuscript is well prepared and clearly structured.

We thank Reviewer #2 for his/her careful and thoughtful review, as well as his/her concise and accurate summary of our work. We fully agree with the Reviewer that the ability to pull absorbing is unique and unexpected. We thank Reviewer #2 for highlighting the novelty, robustness, rigor, and broad interest nature of our theory.

I am pleased to recommend it for publication in Nature Communications, and I have the following minor comments for the authors' consideration.

We thank Reviewer #2 for recommending the publication of our work in Nature Communications. The concerns raised are answered point-by-point below.

### 1. The authors have studied the pulling force on dielectric particles in detail. Does the mechanism apply to metallic particles of arbitrary size?

Our analytic derivation is valid for an arbitrary particle, irrespective of its size, shape, and composition (can be absorbing). So indeed, the mechanism applies to metallic particles of arbitrary size and shape. In the newly added Fig. 5f, the red line explicitly illustrates the pulling force acting on a metallic particle with a dielectric constant -3+0.5*i*, while other curves plotted the pulling force acting on particles with different compositions and morphology.

3. The lateral optical forces also strongly depend on the surface waves or guided waves of the substrate. Can the mechanism be applied to reverse the lateral optical force?

We thank Reviewer #2 for drawing us to this subtle issue.

As discussed in the main text, the lateral optical force  $F_y$  has a similar expression as Eq. (6):

$$
F_{y} = -\frac{1}{v_{p}} W_{sca}^{(s)} \langle \cos \phi \rangle - \frac{1}{c} W_{sca}^{(f,a)} \langle \cos \theta \rangle_{1} - \frac{1}{v_{f}} W_{sca}^{(f,m)} \langle \cos \theta \rangle_{2}, (R2)
$$

where  $\langle \cos \phi \rangle$ ,  $\langle \cos \vartheta \rangle_1$  and  $\langle \cos \vartheta \rangle_2$  are the averaged cosines of the scattering angles measured with respect to the *y* axis (not *x* axis as in the main text). According to Eq. (R2), the term associated with incident optical force, which is related to the extinction rate, is absent. As a consequence, replacing the incident SW with a positive propagation constant  $(k\,p)$  by a SW with a negative k p does not guarantee the reversal of the lateral optical force, because there is no reversal of the incident force.

However, the scattering force related to the scattered SWs (the first term in Eq. (R2)) is reversed when the sign of v\_p is altered. Additionally, if a SW with negative k\_p is supported, the sign of v\_f of the metamaterial is also altered in general, resulting in the reversal of the scattering force related to the FPWs inside the metamaterial (third term in Eq. (R2)). In summary, our mechanism can be applied to reverse parts of the lateral optical force, thus favoring complete reversal in most cases, except when the incident SW is mainly scattered into the FPWs in the air.

4. In experiments, there are usually many particles on the substrate. Will the pulling force be affected by the multiple scattering of the particles?

#### We thank Reviewer #2 for drawing us to this important issue.

For the simultaneous pulling of multi-particles, our analytical theory guarantees a negative total force (because the entire cluster can be considered as a single particle), but there is no guarantee that the forces on individual particles are negative. Nevertheless, the momentum absorbed by all particles is always negative. Thus, the whole system is always being pulled, and all particles will eventually be pulled back to the surface wave source without exception. As shown in newly added Supplementary Figure 6, three particles with different shapes and electromagnetic properties are considered. In panels a-c, all three particles experience pulling forces regardless of their separations. In panel d, where the second particle is metallic, the longitudinal optical force on the second particle may become positive for certain distances. Nevertheless, the total longitudinal optical force acting on the three particles remains negative.



**Supplementary Figure 6**. The longitudinal optical forces acting on three particles with particle sizes **a**  $k_0r=0.5$ , **b** and **d**  $k_0r=1.5$ , **c**  $k_0r=2.5$ . The inset in panel **c** illustrates the arrangement of the particles, equidistant along the *x*-direction. The first and third particles are lossless ( $\varepsilon_r = 5$ ,  $\chi = 0$ ) and chiral ( $\varepsilon_r$ =5,  $\chi$ =1.0), respectively. In panels (**a-c**), the relative permittivity of the middle particle is ε*r*=5+0.5*i*, while in panel **d** it is ε*r*=-3+0.5*i*.

#### **REVIEWER COMMENTS**

#### **Reviewer #1 (Remarks to the Author):**

I have found that the authors of this article have improved the quality of the article significantly. Notably I refer to the addition of figure 5 in their article along with other important revisions. However, a few notable revisions are still highly recommended:

1. The quality of the rebuttal letter is very nice. But only a portion of that letter has been reflected in the main article. I suggest the authors clarify the equation of the incident beam given in Figure 5. Without such an equation, how this or similar work will be reproduced by the other groups citing this article!!! Reproducibility is an important aspect of good scientific work. Such an equation should be put in their newly added section: Optical pulling over a realistic metamaterial structure. Also I suggest the authors properly mention the value of the amplitude of the incident beam (along with the polarization).Also authors should discuss the 45 degree angle issue in the main article (which they have discussed in the rebuttal letter).

2. The structure presented in Figure 5 is very similar to the structures reported previously in "Mode conversion enables optical pulling force in photonic crystal waveguides." Applied Physics Letters 111, no. 6 (2017)." and "Optical pulling using evanescent mode in subwavelength channels." Optics express 24, no. 16 (2016): 18436-18444.

I humbly suggest the authors clarify the difference of their optical pulling comparing aforementioned tractor beam type effects in APL and OE. Othwersie, even if this work gets published (in future), experts will certainly point to this notable similarity.As a result, a proper explanation (both in the rebuttal letter and main article) is highly recommended regarding this matter.

3. The issue of loss (metamaterial) and its impact has been given in the supplement section (supplement note 7). A short discussion [some numerical values of loss; permittivity] on this matter can be given in the main article.

## **Reviewer #2 (Remarks to the Author):**

The authors have well addressed my comments. They have added new results to further strengthen the theory. I believe this work will attract broad interest. I am very happy to recommend this revised version for publication.

## **Response to Referees**

#### **Reviewer #1 (Remarks to the Author):**

I have found that the authors of this article have improved the quality of the article significantly. Notably I refer to the addition of figure 5 in their article along with other important revisions. However, a few notable revisions are still highly recommended:

We thank Reviewer #1 for his/her careful and continuous review, as well as for recognizing our effort in improving the manuscript. The recommendations of Reviewer #1 are addressed one by one below.

1. The quality of the rebuttal letter is very nice. But only a portion of that letter has been reflected in the main article. I suggest the authors clarify the equation of the incident beam given in Figure 5. Without such an equation, how this or similar work will be reproduced by the other groups citing this article!!! Reproducibility is an important aspect of good scientific work. Such an equation should be put in their newly added section: Optical pulling over a realistic metamaterial structure.

We thank the reviewer for raising this important issue. The incident beam profile is indeed very important, and it is our responsibility to specify the excitation conditions so that interested readers can reproduce our results. Yet, the SW, like other eigenmodes of photonic crystal, has no closedform expression. We now specified in the main text how the SW is generated and plot its profile numerically.

To excite the SW efficiently, phase matching is of paramount importance, while other details of excitation is relatively insignificant. We suggested two methods to phase match the SW for efficient excitation, namely the line sources approach and the prism approach.

For the line sources approach, we have added the following sentences in the third paragraph of the section "optical pulling over a realistic metamaterial structure."

*As shown in Fig. 5b, the line sources, each with an amplitude of 1 mA, are positioned one lattice constant below the metamaterial surface and are equally spaced with a distance a between them. From left to right, the initial phase of the j-th line source is (1-j)×0.2π.*

For the prism approach, the following paragraph is added to the section "optical pulling over a realistic metamaterial structure."

*To excite the SW, it is crucial to phase match its negative k vector. This can be achieved by using a prism (Fig. 5d) or a series of linesources embedded inside the metamaterial (Fig. 5a). Like other eigenmodes of a photonic crystal, there is no closed-form expression for the SW. However, the beam profile has been calculated numerically as shown in Fig. 4c, and an enlarged portion of Fig.5a is also given in supplementary Fig. 4. For prism excitation of the TM-polarized OSTB, an approximate Gaussian incident wave is normally illuminating the right surface of the prism. The incident and prism angle of 45 degrees, and the prism's refractive index of 1.69, are chosen such that the incident wave vector, after entering the prism, can phase match the SW. If the refractive index of the prism differs from 1.69, then other appropriate incident angle can be adopted to phase match the SW and achieving efficient excitation. Following one of the standard approach in* 

*COMSOL Multiphysics finite element simulation, the incident wave is generated by using Gaussian distributed current sources located at the right surface of the prism, i.e.,*   $J_0 \exp(-r_g^2/w_0^2)$ , with its center coincides with the center of the right surface,  $J_0 = 1$ mA/m,  $w_0 = 15a$  and  $r_g$  is the distance from the surface center to the local point on the surface. Then, *the focal plane coincides with the prism's right surface , with beam waist ~15 a and electric field amplitude ~1.37 V/m at the beam focus. In Supplementary Note 5, we explore how the incident angle and polarization of the incident beam affect the excitation efficiency.*

Also I suggest the authors properly mention the value of the amplitude of the incident beam (along with the polarization).Also authors should discuss the 45 degree angle issue in the main article (which they have discussed in the rebuttal letter).

The value of the amplitude of the incident electric field is  $\sim$  1.37 V/m at the focus of the beam, and the incident beam is TM polarized. The incident and prism angle of 45 degrees, together with the refractive index of 1.69 for the prism, are chosen such that the incident wave vector, after entering the prism, can phase match the SW. If the refractive index of the prism differs from 1.69, then other appropriate incident angle can be adopted to phase match the SW and achieving efficient excitation. That information has been added to the main text through the paragraph given above.

2. The structure presented in Figure 5 is very similar to the structures reported previously in "Mode conversion enables optical pulling force in photonic crystal waveguides." Applied Physics Letters 111, no. 6 (2017)." and "Optical pulling using evanescent mode in sub-wavelength channels." Optics express 24, no. 16 (2016): 18436-18444. I humbly suggest the authors clarify the difference of their optical pulling comparing aforementioned tractor beam type effects in APL and OE. Othwersie, even if this work gets published (in future), experts will certainly point to this notable similarity.As a result, a proper explanation (both in the rebuttal letter and main article) is highly recommended regarding this matter.

We thank the reviewer for pointing this out. The following paragraph is added to the section "Optical pulling over a realistic metamaterial structure" of the main text.

*Previous studies, such as references 38 and 68, have demonstrated the use of photonic crystal structures to induce optical pulling forces. However, our approach is fundamentally different. Due to the lattice constant being much smaller than the wavelength, the photonic crystal we employ functions effectively as a double-negative-index metamaterial. This characteristic allows the SW to be well described by a single negative Bloch wavevector. This unique property of the SW enables a dominant negative incident force, ensuring the optical pulling of arbitrary particles along the surface of the photonic crystal structure. Unlike previous works [38, 68], our approach does not rely on any directional waveguide mode conversions, eliminating the need to bound the particle inside a photonic crystal.*

3. The issue of loss (metamaterial) and its impact has been given in the supplement section

(supplement note 7). A short discussion [some numerical values of loss; permittivity] on this matter can be given in the main article.

We thank the reviewer for his/her valuable suggestion. In the main text (first paragraph of discussion section), we have added the following sentences to address the issue of loss:

*As shown in Supplementary Fig. 7, when the relative permittivity and permeability of the metamaterial become*  $\varepsilon = -0.9 + 0.02i$ ,  $\mu = -1.2 + 0.02i$ , the particle continues to experience a *pulling force. However, since the OSTB decays as it propagates, the optical pulling range will be shortened.*

# **Reviewer #2 (Remarks to the Author):**

The authors have well addressed my comments. They have added new results to further strengthen the theory. I believe this work will attract broad interest. I am very happy to recommend this revised version for publication.

We thank Reviewer #2 for his/her careful and in-depth review, which has significantly improved our manuscript.