

Negative force on free carriers in positive index nanoparticles

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(Dated: 8 August 2017)

We theoretically demonstrate the reversal of optical forces on free charge carriers in positive refractive index nanostructures. Though optical momentum in positive refractive index materials is necessarily parallel to the local energy flow, reversal of optical momentum transfer can be accomplished by exploiting the geometry and size of subwavelength particles. Using the Mie scattering theory and separation of optical momentum transfers to the bound and free charges and currents, we have shown that metal nanoparticles can exhibit strong momentum transfer to free carriers opposite to the direction of incident electromagnetic waves. This can be explained for small particles in terms of a reversal of Poynting power inside the material resulting in a negative net force on free carriers in small particles. Two-dimensional simulations further illuminate this point by demonstrating the effect of incident wave polarization.

PACS numbers: Valid PACS appear here

The reversal of radiation pressure in negative index materials (NIMs) was first predicted by the seminal work of Veselago¹. NIMs have received significant attention since their realization in 2001² due to the potential for interesting physics such as reversal of the Cerenkov effect³, perfect lensing⁴, and negative refraction⁵. Veselago's results predict in materials with simultaneously negative permeability and permittivity ($\mu < 0$, $\epsilon < 0$), light attraction instead of light pressure, and momentum transmitted into an NIM is antiparallel to the Poynting power. The radiation force on free carriers (F_c) in a material becomes reversed if the index of refraction is negative ($n < 0$) due to the reversal of wave momentum⁶.

J. C. Maxwell first applied the electromagnetic wave theory to predict and calculate the radiation pressure of light⁷. Observation of light pressure on reflectors in vacuum validated this prediction^{8,9}. After, detailed quantitative measurement of radiation pressure was presented by Poynting in 1905¹⁰. First observed in 1970, the photon drag effect results from the optical momentum transfer in semiconductors such as Germanium or silicon^{11,12}. The experiment reveals that when a photon of energy \mathcal{E} is absorbed in a semiconductor of refractive index n , the Minkowski momentum $n\mathcal{E}/c$ is transferred to free carriers, where c is the vacuum speed of light. From the momentum conservation, additional force on the bulk material must be directed toward the incident wave. Loudon *et al*¹³ provided a theoretical description of momentum transfer from electromagnetic wave to charge carriers and host semiconductor for limiting cases introducing the division of momentum transfers to a force on free carriers and a force on the host or bound carriers.

In 1954, Jones and Richards found the radiation pressure exerted upon a submerged mirror is proportional to the index of reflection n of the surrounding medium by measuring the deflection of refractors in submerging fluids¹⁴. This experiment failed to determine whether

the proportionality of refraction index was phase velocity $n = c/v_p = ck/\omega$ refractive index or the group velocity refractive index $n_g = c/v_g = c\partial k/\partial\omega$, where v_p and v_g are the phase and group velocities, respectively, ω is the frequency, and k is the wavenumber. Jones and Leslie repeated the original experiment with the use of a laser in 1978 and determined that the pressure upon a mirror submerged in a dielectric liquid is proportional to the phase velocity refractive index n ¹⁵.

In 2005, Campbell *et al*, presented a measurement of photon recoil momentum of single atoms in a dispersive medium consisting of a gas of ultra-cold atoms¹⁶. The experiment revealed the change in momentum of single atoms in the lowest momentum state caused by absorption of photons. It was observed using an atom interferometer that the recoil via optical momentum transferred to the atoms was directly proportional to the classical, macroscopic refractive index n of the gas. The experiment also shows that the presence of all surrounding atoms affect the single photon entering a dilute gas.

In 2015, Tengfei Zhang *et al*. group demonstrated remarkable light induced propulsion and rotation of bulk graphene based material¹⁷. It is stated that the unique structure and properties of the graphene sponge make it not only absorb light, but it also can emit high velocity electrons which have net momentum towards the light source. This net electron momentum driven toward the laser source can propel the material along the propagation direction of the laser at a momentum larger than the incident photons. Although the incident optical momentum is positive in this case, the net momentum of ejected electrons towards the light source, which facilitates the propulsion, was experimentally shown to be negative. According to the previous discussion, the force on free carriers should be positive in a medium with positive real part of the refractive index $n = n_R + in_I$. In this experiment, the observed electron momentum is negative. However, the experiment has been presented with an energy analysis, but description as to the mechanism for momentum transfer to the electrons yielding negative momentum. A recent thermal analysis questioned the

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physical mechanism responsible for the propulsion of the graphene sponge as radiometric forces may be responsible¹⁸. However, the debate is still ongoing as to the description of this phenomenon¹⁹.

In this Letter, we theoretically demonstrate that force on free carriers can be negative in a positive refractive index medium. It is shown that such negative momentum transfer to free carriers accompanies enhanced positive forces on the host material (i.e. bound carriers). We demonstrate this using metal particles with ($n_R > 0$). Our conclusions reveal that the negative momentum transfer free carriers within positive refractive index media is highly dependent upon the sub-wavelength geometry of the absorber. The justification for our application to metal particles is that it provides a proof-of-concept rooted in well-known material systems with analytical solutions. Whereas application to specific experimental conditions, as we point out, will require very detailed information about the sub-micron geometries involved. For example, we have calculated the optical forces on bound and free carriers for gold and silver nanospheres directly with complex permittivity $\epsilon(\omega) = \epsilon_R + i\epsilon_I = |\epsilon| \exp(i\phi_\epsilon)$ and permeability $\mu(\omega) = \mu_R + i\mu_I = |\mu| \exp(i\phi_\mu)$ in vacuum (ϵ_0, μ_0). The total time-averaged optical force $\bar{F} = \bar{F}_b + \bar{F}_c$ due to $e^{-i\omega t}$ dependent harmonic excitation is decomposed into the force on bound carriers \bar{F}_b and force on free carriers \bar{F}_c based on the real and imaginary contribution to the complex permittivity and permeability¹³. The same approach has been previously been previously applied to model the momentum transfer to the isotropic left handed media⁶ and absorbing Mie particles²⁰.

The momentum of light in media and the resulting forces is the subject of a longstanding debate, and attention must be given to the formulation of electrodynamics applied in the theoretical model²¹⁻²³. We begin with the field-kinetic stress-energy-momentum tensor resulting from the Chu formulation, which is interpreted as providing the action of the field energy and momentum upon media^{24,25}. The force density^{6,20}

$$\bar{f}_c = \frac{1}{2} \Re \left\{ \omega \epsilon_I \bar{E} \times \mu_0 \bar{H}^* - \omega \mu_I \bar{H} \times \epsilon_0 \bar{E}^* \right\}, \quad (1)$$

provides the field-kinetic momentum transfer to free carriers as the wave attenuates in the medium. By applying Poynting's theorem, the force density on free currents can be shown to be written as $\bar{f}_c = -\hat{z} \frac{1}{2} \text{Re} \left\{ \frac{n}{c} \bar{\nabla} \cdot \bar{S} \right\}$ where $\bar{S} = \bar{E} \times \bar{H}^*$ is the complex Poynting power²⁰. Thus, the direction of the force on free currents depends upon the sign of the index of refraction n . Additionally, the force density on bound currents and charges

$$\bar{f}_b = \frac{1}{2} \Re \left\{ (\nabla \cdot \bar{P}) \bar{E}^* + (\nabla \cdot \mu_0 \bar{M}) \bar{H}^* - i\omega(\epsilon_R - \epsilon_0) \bar{E} \times \mu_0 \bar{H}^* + i\omega(\mu_R - \mu_0) \bar{H} \times \epsilon_0 \bar{E}^* \right\}, \quad (2)$$

provides the remaining momentum transfer to the host material. The total force on a material \bar{F} and the total force on free \bar{F}_c and bound \bar{F}_b carriers result from the integration of the force densities over the entire medium.

The Chu force on a particle can be written as below^{6,20,26}

$$\bar{f}_{ch} = \rho_e \bar{E} + \rho_m \bar{H} + \bar{J}_e \times \mu_0 \bar{H} - \bar{J}_m \times \epsilon_0 \bar{E} \quad (3)$$

where $\rho_e = -\nabla \cdot \bar{P} + \rho$ and $\bar{J}_e = \frac{\partial \bar{P}}{\partial t} + \bar{J}$. So for the non-magnetic particle Chu force can be written as below

$$\bar{f}_{ch} = \rho \bar{E} + \bar{J} \times \mu_0 \bar{H} + (-\nabla \cdot \bar{P}) \bar{E} + \frac{\partial \bar{P}}{\partial t} \times \mu_0 \bar{H} \quad (4)$$

From the definition $(-\nabla \cdot \bar{P}) \bar{E} + \frac{\partial \bar{P}}{\partial t} \times \mu_0 \bar{H}$ is the force on bound carriers and charges, \bar{f}_b . So the force on free carriers and charges becomes

$$\bar{f}_c = \rho \bar{E} + \bar{J} \times \mu_0 \bar{H} \quad (5)$$

Now from the Minkowski formula the force on a non-magnetic particle is^{21,26}

$$\bar{f}_m = -\frac{1}{2} (\bar{E} \cdot \bar{E}) \nabla \epsilon + \rho \bar{E} + \bar{J} \times \mu_0 \bar{H} \quad (6)$$

When we integrate outside the particle we get total force but when we integrate just inside the homogeneous particle to find the force of free carriers and charges and in this case, $\nabla \epsilon$ becomes zero. So the $\bar{f}_c = \rho \bar{E} + \bar{J} \times \mu_0 \bar{H}$ for Minkowski formulation also gives the same formulation as Chu which is shown in Eq. (5) and this is consistent with the Lorentz force on free carriers. So, the calculated force density \bar{f}_c is identical between the field-kinetic (Chu) and canonical (Minkowski) formulations, whereas the distribution of \bar{f}_b differs²¹. However, the total values for both \bar{F}_c and \bar{F}_b will be identical due to the necessity of resulting calculations for \bar{F} being identical²⁶

The connection of momentum transfer to bound and free currents is formalized here by applying the momentum conservation theorem via the Maxwell stress tensor. The time-average force on all currents and charges in a volume V enclosed by a surface of area A with area element $\bar{d}a$ can be determined by applying the divergence of the Maxwell (vacuum) stress tensor. For distinguishing the values of \bar{F}_c and \bar{F}_b , we may apply the complex Minkowski stress tensor

$$\bar{T} = \frac{1}{2} (\bar{D} \cdot \bar{E}^* + \bar{B}^* \cdot \bar{H}) \bar{I} - \bar{D} \bar{E}^* - \bar{B}^* \bar{H} \quad (7)$$

to calculate the momentum transfer to free carriers enclosed by a surface of area A within a homogeneous medium

$$\bar{F} = -\frac{1}{2} \Re \left\{ \oint_A \bar{d}a \cdot \bar{T}(\bar{r}) \right\}. \quad (8)$$

Therefore, the force on free carriers \bar{F}_c is found by integrating the Minkowski stress tensor just inside the surface, and the total force \bar{F} is calculated by integrating the stress tensor outside the particle surface²⁰. The force on the host (i.e. bound carriers) is determined, then, by $\bar{F}_b = \bar{F} - \bar{F}_c$.

The total fields presented herein are due to a plane wave $\bar{E}_{inc} = \hat{x} E_0 e^{ik_0 z}$ incident from free space and are found using Mie theory²⁶⁻²⁸. Because the incident power and momentum are \hat{z} directed, we consider the z components of the forces, which we denote simply as F, F_b

and F_c , where a negative force indicates the $-\hat{z}$ direction. The dielectric constant of metal particles with radii, less than 10 nm is size dependent because in this size range the radius of the particle is comparable to the conduction electron mean free path²⁹. It is not our intent, nor is this effect dependent upon quantum size effects of the nanoparticles or nanorods. As such, we have removed mention of particles less than 10nm from the calculation of optical force.

Fig. 1 shows the separation of total Lorentz force, F (black line) into F_b (dashed line) and F_c (double dashed line) for gold nanoparticles with size range, $a = 10 - 30nm$. Gold nanoparticles have frequency dependent permittivity and which can be negative in some frequency range but the index of refraction remains positive. Here gold has permittivity, $\epsilon = -5.6141 + 2.2556i$ at $\lambda = 532nm$ with positive refractive index $n = \sqrt{\epsilon_r \mu_r} = 0.467 + 2.415i$ ^{30,31}. In this case, the F_c gives negativity with a positive index of refraction for the gold nanoparticle. However, the F_c is positive and F_b is negative for absorbing particle with a similar size range of particle when the real part of permittivity is positive with a positive index of refraction (data not shown). So, enhancement of F_b is possible for metal particles due to the negative force of free carrier and charges. For example at $a = 15nm$, $F_c = -1.6575 \times 10^{-19}nN$ and $F = 9.1624 \times 10^{-19}nN$, so $F_b = F - F_c = 1.0820 \times 10^{-18}nN$. We believe even stronger F_b is possible with a different parameter which can provide large propulsive force to the material.

The negative F_c depends on the particle's size and geometry. Double dashed line in the inset of Fig. 1 shows zoomed out plot of F_c for gold nanoparticle with a positive refractive index using the same parameters. The F_c is minimum when the size of gold particle is 32nm and becomes positive at 42 nm for gold. Similar results are also found for other materials (e.g. silver), although the results are omitted for brevity.

We further investigate the reason for the negativity of F_c by plotting real part of Poynting power in the x-z plane. According to the Eq. (1), the F_c depends on the real part of Poynting power. Fig. 2 shows the direction of real part of complex Poynting power \bar{S} for gold nanoparticle using the same parameters of Fig. 1 at $a = 20nm$ when the F_c is negative. It is observed that real part of S becomes negative at the inside of the material and gives negative F_c . The derivation of $\bar{f}_c = -\hat{z}\frac{1}{2}Re\{\frac{n}{c}\bar{\nabla} \cdot \bar{S}\}$ from Ref.²⁰ assumes a plane wave solution as a uniform field amplitude. This condition is met in Rayleigh regime as seen in Fig. 2, but not when the diameter is larger than $\lambda/10$. Furthermore, $F_c < 0$ in the Rayleigh regime when the Poynting power in reversed. When the size parameter for the particle is larger than the Rayleigh regime this effect is not observed. From the Eq. (2), the F_b depends on the imaginary part of Poynting power. It is also observed that imaginary part of \bar{S} is strongly negative inside the material and gives strong F_b when F_c is negative. Negative F_c is only found at the off-resonance condition. At resonance condition of gold and silver, the force is stronger but no negative F_c is observed (data not shown). We have also shown the frequency dependency of negative force on free carriers and charges, F_c in Fig.

3. We have determined separation of total Lorentz F (solid line) into F_b (dashed line) and F_c (double dashed line) for a gold sphere of radii, $a = 20nm$ for the wavelength range, $\lambda_0 = 200 - 700nm$. Wavelength dependent dielectric constant is used from tabular data³⁰. Gold particle shows resonance at the scattering light, $\lambda_0 = 481nm$ when $Re(\epsilon) = -2$ ^{30,32}. It is seen that the F_c is negative for a wide frequency range due to the reversal of Poynting power, \bar{S} inside the material when it crosses the resonance frequency. For the silver particle similar effects has also found (data not shown).

For completeness, that F_c can be negative in metal nanostructures besides the spherical metal nanoparticles we have also calculated the separation of total Lorentz F (solid line) into F_b (dashed line) and F_c (double dashed line) for infinite gold cylinder using the same parameters of Fig. 1. We have used normalized force in the results of the infinite cylinder by using incident momentum, $\langle P_0 \rangle = \langle S_0 \rangle / c = 1$. Fig. 4 and Fig. 5 show the incident wave polarization dependency of F_c for both TE and TM modes. For TE mode of propagation, the \bar{E} is polarized along the infinite axis of the cylinder, and there is no negative F_c found as shown in Fig. 4. But for the case of TM mode of propagation, the \bar{E} is polarized along the radius of the cylinder, and the F_c shows similar negativity as the spherical particle which is shown in Fig. 5. The size dependency as like spherical particle is also observed in an infinite cylinder.

In summary, the F_c can be negative in positive index absorbing nanoparticles which depends on the size of that nanoparticles. We also demonstrate the polarization effects with infinite cylinders, which indicate a geometric effect along the size effect for realizing negative optical momentum transfer to free carriers in positive refractive index media. If the free carriers come out from the particle then the total force on the particle is just the F_b and when the F_c is negative, by tuning the parameters, polarization and geometry of particles useful strong F_b can be achieved to gain propulsive force¹⁷. The reversal of F_c is also can be an indirect explanation for the negative momentum of electron towards a light source and strong propulsive force to the host material. Whether the free carriers come out from the particle or remain in the particle depends on the momentum of incoming light which can be said as the photoelectric effect in metal. Negative F_c results can be verified into a (near) vacuum to avoid the probable interference of air when the free carriers come out from the particle¹⁹. Although there is a lot of research on metal particle/positive index media, but to the best knowledge of authors, the negative force on free carriers is not measured for the positive index of media yet except for the graphene. Our results are theoretical, and we expect additional experimental results similar to the graphene those published by T. Zhang et al. in 2015 to verify the model with additional material systems in future.

Funding for this research was provided by the Center for Advanced Surface Engineering, under the National Science Foundation Grant No. IIA-1457888 and the Arkansas EPSCoR Program, ASSET III and by the NSF Division of Electrical, Communications, and Cyber Systems (Award No. ECCS-1150514) through the

Faculty Early Career Development (CAREER) program. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the National Science Foundation.

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- FIG. 1. Forces vs. radius for gold sphere ($\epsilon = -5.6141 + 2.2556i$ and $n = 0.467 + 2.415i$) incident by a unit amplitude plane wave. The free space incident wave is $\lambda_0 = 532nm$ and number of modes, $N = 20$. Zoomed out plot of forces on free carrier and charges, F_c vs radius for gold sphere using same parameters is shown in inset.
- FIG. 2. Real part of Poynting power, $S_R = \Re\{E \times H^*\}$ on a $20nm$ radius gold sphere ($\epsilon = -5.6141 + 2.2556i$ and $n = 0.467 + 2.415i$) incident by a \hat{x} polarized unit amplitude plane wave. The free space incident wave is $\lambda_0 = 532nm$ and number of modes, $N = 20$.
- FIG. 3. Forces vs. wavelength for a gold sphere of radii, $a = 20nm$ and incident by a unit amplitude plane wave. The free space incident wave is $\lambda_0 = 200 - 700nm$ and number of modes, $N = 20$.
- FIG. 4. Normalized Forces vs. radius for infinite gold cylinder ($\epsilon = -5.6141 + 2.2556i$ and $n = 0.467 + 2.415i$) of radii, a and incident by a TE mode unit amplitude plane wave. The free space incident wave is $\lambda_0 = 532nm$ and number of modes, $N = 20$.
- FIG. 5. Normalized Forces vs. radius for infinite gold cylinder ($\epsilon = -5.6141 + 2.2556i$ and $n = 0.467 + 2.415i$) of radii, a and incident by a TM mode unit amplitude plane wave. The free space incident wave is $\lambda_0 = 532nm$ and number of modes, $N = 20$.









