

Symmetry Breaking-Induced N-body Electrodynamic Forces in Optical Matter Systems

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The research on "Symmetry Breaking-Induced N-body Electrodynamic Forces in Optical Matter Systems" provides a compelling and innovative exploration into the realm of non-reciprocal forces within optical matter systems. John Parker et al., addresses a significant gap in the understanding of N-body non-reciprocal forces induced by symmetry breaking. By focusing on systems with three or more nanoparticles, it goes beyond the typical pairwise interactions explored in previous studies. Moreover, the combination of experimental work, numerical simulations, and theoretical analysis provides a robust framework for investigating the phenomena. The use of optical ring traps to sustain the bent configuration of nanoparticles is particularly innovative, ensuring the stability of the bent structures necessary for observing N-body forces. The discovery of N-body non-reciprocal forces in optical matter systems has broad implications. It opens new avenues for the design of advanced materials and devices that leverage these unique interactions, potentially impacting fields such as photonics, material science, and active matter physics. Therefore, I recommend its publication in Nature Communications. The minor suggestions provided aim to further refine the manuscript and enhance its impact.

1. While the use of optical ring traps is innovative, the practical implementation of these setups can be complex and may require significant precision. The paper does not extensively discuss potential limitations or challenges in replicating these experiments in different settings.
2. The study primarily focuses on systems with Ag nanoparticles. It would be beneficial to see how these findings translate to other materials or nanoparticle configurations, potentially broadening the applicability of the results.
3. The stability of the observed configurations over extended periods, especially under varying environmental conditions, is not fully addressed. This could be an important factor for practical applications of these findings.
4. While it is possible to impose negative torques on optical matters via a single plane-wave, which depends on the specific configuration of the particles, does the nanoparticles-in-the-ring exhibit negative optical torque?

(Remarks on code availability)

Reviewer #2

(Remarks to the Author)

In this work, the authors theoretically point out and demonstrate numerically and experimentally that symmetry breaking may give rise to N-body non-reciprocal interactions ($N \geq 3$) even when the particles are identical (unlike in the majority of the works in the literature that consider two-body non-reciprocal interactions for different types of agents). Using optically trapped Ag nanoparticles with external circularly polarized light injection, the authors demonstrated that the light-scattering induces a tilt of the dipoles that induces a flux of electromagnetic field, giving rise to a net force of the nanoparticles. The observation that the symmetry-breaking induces a non-reciprocal interaction for a multi-body interaction is very interesting and novel. The data and the comparison between numerics and experiments are convincing. I recommend the manuscript to be published in Nature Communications once the comments below are addressed.

(1) In my understanding, in the two-body case with NON-identical nanoparticles (as some of the authors have demonstrated in their previous works), the presence of dissipation played a key role in obtaining non-reciprocity. That is, when light is injected to the two nanoparticles A and B, the light that scatters from A (B) propagates to optical force on B (A) to give rise to forces acting between the two nanoparticles. When A and B have different absorption rates, the amount of light scattering is different, giving rise to non-reciprocal interactions. Am I right that this situation (i.e. the absorption plays a crucial role for non-reciprocity to emerge) holds true for the N-body non-reciprocal interaction demonstrated in this work? If so, I believe it would be helpful to emphasize a little more explicitly (probably in the first paragraph of the “Results” section) that absorption plays a crucial role. If not, it would be interesting to add a discussion on this intriguing difference.

(2) (Slightly related to above) I was confused with the definition of α . Is α a complex number including absorption or a real number? I got confused because, in Eq. (2), the second term is proportional to α^* (with *) implying α includes an imaginary part (which physically makes sense to me), while in Fig. 2(b), α is assigned with a real number. Please clarify the definition.

(3) Below Eq. (2), it would be helpful for the readers if they remarked that the second term vanishes because the particle is identical.

(4) The authors use the words ‘fully filled’ ‘half filled’ etc. as if there is an underlying lattice structure (such that a filled or vacant site can be defined). To my understanding, the crystalline structure is emergent due to the reciprocal piece of interaction and is not something that the authors externally imposed. It would be helpful to clarify this point in the manuscript, as the definition of ‘fully filled’ etc. was not so clear to me.

(Remarks on code availability)

Reviewer #3

(Remarks to the Author)

The authors present interesting results on the non-reciprocal forces in a N-body system that induces active motion of a cluster of nanoparticles confined within an annular optical beam. They show that due to the optical confinement of the Ag particles on a ring their individual scattering leads to a breaking of the symmetry between each and other and overall a net force that creates circular motion depending on the polarisation of the trapping beams.

The experimental results are sound and clearly supported by rigorous simulations. Great care has been taken to ensure that no other optical effects such as aberrations to interfere with the induction of a net force. Whether the basis of the simulations are reasonable for such experimental system is outside my scope of expertise.

However, the current version of the manuscript does not fully justify the claims of the paper on symmetry breaking of N-body systems in optical matter systems. Although the authors clearly demonstrate N-body forces within their experimental system, there are already several examples of such systems, also indicated by the references provided. A quick literature research showed up other examples of an optical system (Thermal-motion-induced non-reciprocal quantum optical system, Nonadditivity of critical Casimir forces) where symmetry is being broken and lead to the creation of additional forces in the system. A clearer justification within the context of the present literature is required to better support the new findings the authors try to claim.

In its current state, I fail to see the significant advancements of our understanding on non-reciprocal to the field of, e.g., active matter.

Overall, the manuscript requires significant restructuring and modification of the introduction, results and discussion section to provide better readability and enhancing the clarity of the presented results. A large part of the current introduction has been dedicated to the results of the paper while the literature has been only briefly mentioned. Here, a larger introduction into the topic of non-reciprocal forces would greatly benefit the reader to place the authors work into context. Also why symmetry and the breaking of it are important, for instance in the field of active matter, is lacking in further depth. This should allow the reader to better understand how the results could be used in different communities. A further description of the current limitations of other studies on N-body forces, and how the authors work tackles those is missing as well. In the beginning of the results section a flow from the context of the literature into the experimental system should be provided. Here a brief description of the experimental system and other parameters used would provide a basis for the results presented later. Without it, as a reader, one is quickly lost in the details of the results and requires significant forth and back between the description of figures and crucially missing info in written either in the methods section or hidden in the Supplementary Information. More clear referencing from the main manuscript to the SI is needed to allow for quick referencing. In contrast to the introduction, the discussion section recites again the literature and challenges, explaining only here the significance of their work. This should be placed instead in the introduction section. This space should be dedicated better to the discussion of the results, other aspects to be considered, such as the influence of radius of the optical beam. Detailed questions that could partly be answered here are provided below.

Major comments:

1. The authors have used Ag nanoparticles for their study. Could they better justify the choice of such material, metallic vs. dielectric particles (commonly used in experiments), and why Ag in particular?
2. What other forces could be at play in such systems that could drive particle motion besides optical gradients? What about thermal forces due to absorption of light on nanoparticles? For example: <https://www.nature.com/articles/s41467-022-28212-z>
3. From Figure 1h, it is not clear to me why there is only a force component along x here and not y as well. Shouldn't the force be along azimuthal as presented later on as well?

4. Have the authors analysed the trapping stiffness of the optical trap along the radial coordinate?
5. Line 94: What does quickly restored refer to here? Can they provide a number?
6. The authors provide only experimental data for filled rings of two radii and it three configurations. To support their claim and simulations it would be beneficially to also show experimental data of lower number of particles N.
7. Figure 2c: What is the reason for the bent in the graph just above 20 particles? What is the dependence on the distance between particles here for cases where particles are not fully filling the ring?
8. Figure 2d: Why are the distributions of the different sizes here?
9. What was the reasoning behind not using the same power density for the simulations in order to make the comparison with experiments?
10. SI 3: The statement for the step-like behaviour in Fig. S2 would benefit from a more clear illustration of when particles had to be added depending on the ratio between ring and particle diameter.

Minor comments:

Line 41: Here N-body ED forces are immediately assumed which is not obvious to the reader. A better clarification is required.

Line 50: Not clear what bent configuration refers to here.

Line 65: Linear multimers are not shown here, so why mention it?

Line 73: Justification for that statement missing.

Figure 1d/S1: Why are the orientations of the elliptical polarisations not the same? Is that on purpose or an illustration error?

Figure 1g: Not clear to everyone what h.o. stands for, better to use same annotation as in Eqn 2 throughout whole paper.

Figure 1e,f scale bars missing.

Line 97: Not clear what x_e stands for.

Line 123: spacing error

Figure 2a: Missing color bar

Figure 2b: F_{ϕ} should be noted here as variable as throughout the rest of the manuscript. Why the choice of 70x? A better way of presenting the data should be chosen without arbitrary scalings such as normalisations of the curves.

Figure 2 caption: Weird notation of diameter, better way $d=150\text{nm}$, Not consistent notation of time intervals between figure and caption.

Fig. S9: Would be good to see some error bars as individual trajectories should vary quite significantly as indicated by large distributions in the velocities before.

Fig. S12 scale bars not clearly visible.

(Remarks on code availability)

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have thoroughly addressed the reviewers' comments. I recommend the manuscript for publication.

(Remarks on code availability)

Reviewer #2

(Remarks to the Author)

I am happy with all the responses from the authors. I recommend publication to Nature Communications.

(Remarks on code availability)

Reviewer #3

(Remarks to the Author)

The authors have carefully revised the manuscript and SI to include all comments being made, or have clarified the corresponding aspects in more detail.

Given the much improved version of their introduction, the mentioning of limitations of the system and effects of other parameters such as absorption and choice of material, I can now recommend it for publication.

(Remarks on code availability)

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Response to reviewer comments for manuscript entitled “Symmetry Breaking-Induced N-body Electrodynamic Forces in Optical Matter Systems.”

The authors extend their gratitude to the reviewers for their valuable time and insightful comments and suggestions. The following is a point-by-point response addressing the reviewers' comments and concerns. Changes have been made in both the main text and Supplementary Information (SI) in response to these comments. The manuscript and SI have been revised for improved clarity and completeness. All major edits and changes are highlighted in red text.

Reviewer #1 (Remarks to the Author):

Comment #1: The research on "Symmetry Breaking-Induced N-body Electrodynamic Forces in Optical Matter Systems" provides a compelling and innovative exploration into the realm of non-reciprocal forces within optical matter systems. John Parker et al., addresses a significant gap in the understanding of N-body non-reciprocal forces induced by symmetry breaking. By focusing on systems with three or more nanoparticles, it goes beyond the typical pairwise interactions explored in previous studies. Moreover, the combination of experimental work, numerical simulations, and theoretical analysis provides a robust framework for investigating the phenomena. The use of optical ring traps to sustain the bent configuration of nanoparticles is particularly innovative, ensuring the stability of the bent structures necessary for observing N-body forces. The discovery of N-body non-reciprocal forces in optical matter systems has broad implications. It opens new avenues for the design of advanced materials and devices that leverage these unique interactions, potentially impacting fields such as photonics, material science, and active matter physics. Therefore, I recommend its publication in Nature Communications. The minor suggestions provided aim to further refine the manuscript and enhance its impact.

Response #1: We are grateful for the reviewer’s positive assessment of our manuscript and recognition of the innovative aspects of our research.

Comment #2: While the use of optical ring traps is innovative, the practical implementation of these setups can be complex and may require significant precision. The paper does not extensively discuss potential limitations or challenges in replicating these experiments in different settings.

Response #2: We thank the reviewer’s insightful comment on the potential applications and challenges of applying our findings in various environments. Several sections in the Supplementary Information give a very detailed accounting of the sensitivity of the experiment to optical aberrations and the steps that we have taken to both understand their impact on the results and to document these for the purpose of greater understanding in the broader community. This documentation includes some non-idealities that arose (due to the stochastic nature of the experiment) that we again came to understand and explain. In fact, some of these documented details provided new insights vs. the “perfect” conditions of the simulations.

Our discovery of three-body optical forces significantly complements two-body interactions that – in principle – fall short of explaining the electrodynamics in complex, yet not uncommon, systems of multiple identical particles (perhaps even molecules). Despite their significance and ubiquity in optical matter structures, demonstrating three-body optical forces in even controlled settings is challenging. This difficulty largely stems from the dynamic changes in the symmetrical configurations of optical matter structures due to thermal fluctuations or spontaneous addition/removal of nanoparticles. This complicates the analysis of forces, but also revealed interesting perturbations. Therefore, this manuscript focuses on a clear manifestation of an N-body forces arising from symmetry breaking and methodology — using a ring trap — to enable practical measurement of these N-body forces. The ring trap’s curvature, in which the constituent nanoparticles are confined through dipole (intensity gradient) forces, helps preserve the spatial arrangement

of the nanoparticles, thereby facilitating the observation of the deterministic dynamics of N-body interactions.

Comment #3: The study primarily focuses on systems with Ag nanoparticles. It would be beneficial to see how these findings translate to other materials or nanoparticle configurations, potentially broadening the applicability of the results.

Response #3: We appreciate the reviewer's suggestion to explore the broader applicability of N-body interactions in other materials. Equation 1 in the main text demonstrates that all optically polarizable (nano-)particles are theoretically capable of exhibiting the N-body effect. This equation further reveals that the strength of 3-body optical forces is proportional to the third power of the polarizability of constituent nanoparticles, suggesting that the particles with strong light scattering capabilities exhibit a more pronounced N-body effect. This enhancement occurs because strong scattering increases the interactions with adjacent particles' and induced-polarizations therein, as implied in Eq. 4 of the Methods section. Therefore, consistent with our model, a collection of nanoparticles of any material in which one can create a sufficiently large induced-polarization by the incident and scattered electromagnetic fields is expected to exhibit the N-body effect, with more strongly polarizable materials yielding a more noticeable N-body effect. For example, we expect very similar effects for Au nanoparticles.

To illustrate this concept, we consider two types of nanoparticles as examples: metallic Au and dielectric Si. Figures R1a and R1b plot the net forces exerted on trimers of dia. 150 nm Au and Si nanoparticle materials, respectively, as a function of the trimer's bending angle, θ , ranging from 0 to 60 degree. Apparently, both materials demonstrate nonzero net forces due to 3-body interactions, analogous to those shown for Ag in Fig. 1g of the main text. However, the 3-body optical force — and consequently the N-body effect — is greater for the dia. 150 nm Au nanoparticle for its stronger scattering strength at an excitation wavelength of 800 nm compared to the Si nanoparticle. These findings confirm the applicability of N-body effect across a broad range of materials. However, the force is >5x larger for Au nanoparticles vs. Si.

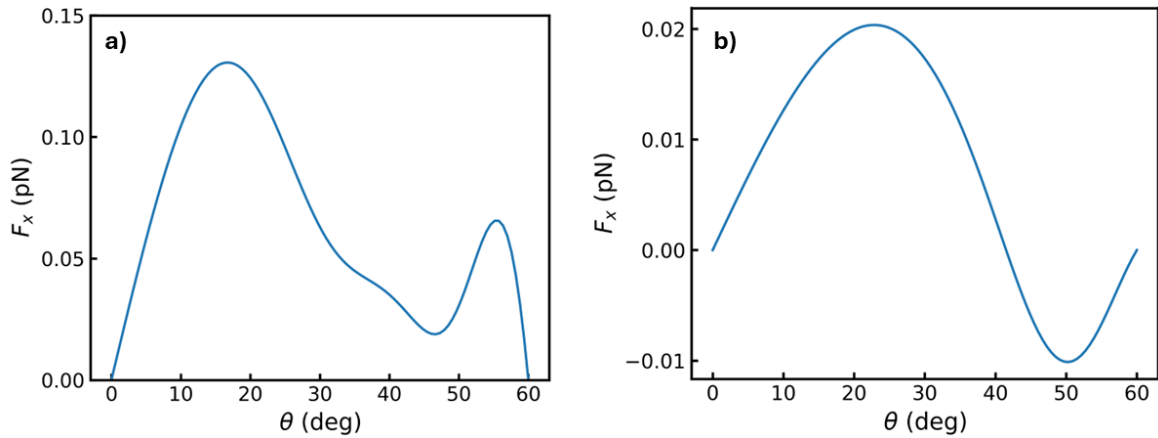


Figure R1. Three-body optical forces of different nanoparticle materials. (a, b) The net forces on trimmer of (a) Au and (b) Si nanoparticles as a function of the trimer's bending angle, θ , for right-handed circularly polarized light at $\lambda = 800$ nm.

Comment #4: The stability of the observed configurations over extended periods, especially under varying environmental conditions, is not fully addressed. This could be an important factor for practical applications of these findings.

Response #4: We thank the reviewer for this question about the stability of ring-configured optical matter structures. We agree that stability is essential for preserving optical matter's structural configurations and, consequently, maintaining their collective dynamics associated with the N-body effect. It is particularly true for two-dimensional (2D) optical matter structures, where experimental perturbations can easily induce structural transitions or particle rearrangement in a less-constrained trapping environment, leading to dynamic changes in their collective electrodynamic responses.

In contrast, the optical matter structures formed in a ring trap are quasi-one-dimensional along the azimuthal direction, with intensity gradient force constraining random (thermal) fluctuations of constituent particles in radial direction. This additional constraint, relative to the formation of 2D optical matter structures, eliminates the likelihood of structural transitions.

However, the performance of ring-configured optical matter structures can still be interrupted by two types of perturbations observed in our experiments: (1) the addition of extra Ag nanoparticles to an already "filled" ring, which may either reduce or increase the N-body forces, and (2) particles becoming "stuck" while in the ring, which could result in transient impedance of collective motions. These perturbing conditions and their consequences have been fully discussed in Section 12 and Section 13 of the Supplementary Information, respectively.

Preventing the addition of unwanted nanoparticles could be achieved with a suitable flow cell in which excess non-trapped particles are removed from the experimental solution volume. Particles do not easily leave the ring trap once they are captured.

Comment #5: While it is possible to impose negative torques on optical matters via a single plane-wave, which depends on the specific configuration of the particles, does the nanoparticles-in-the-ring exhibit negative optical torque?

Response #5: We thank the reviewer's insightful question regarding the possibility of negative optical torques in nanoparticles-in-the-ring configurations. A brief answer is positive based on Fig. R1, where the three-body force of Si nanoparticles at specific bending angles can be negative, indicating the possibility of negative optical torque in such configurations. While we have not yet explored this phenomenon within the scope of the current study, it is an area of interest that we are actively investigating. We value your interest and hope to provide a more comprehensive exploration in a forthcoming project.

Reviewer #2 (Remarks to the Author):

Comment #1: In this work, the authors theoretically point out and demonstrate numerically and experimentally that symmetry breaking may give rise to N-body non-reciprocal interactions ($N \geq 3$) even when the particles are identical (unlike in the majority of the works in the literature that consider two-body non-reciprocal interactions for different types of agents). Using optically trapped Ag nanoparticles with external circularly polarized light injection, the authors demonstrated that the light-scattering induces a tilt of the dipoles that induces a flux of electromagnetic field, giving rise to a net force of the nanoparticles. The observation that the symmetry-breaking induces a non-reciprocal interaction for a multi-body interaction is very interesting and novel. The data and the comparison between numerics and experiments are convincing. I recommend the manuscript to be published in Nature Communications once the comments below are addressed.

Response #1: Thank you for your objective evaluation and insightful comments regarding our manuscript. You pointed out the importance of our work in demonstrating that symmetry breaking can induce non-

reciprocal interactions among identical particles in multi-body systems, a novel direction from prior studies focused on two-body interactions among different agents. We appreciate your recognition of the experimental and numerical demonstrations that underpin our findings.

Comment #2: In my understanding, in the two-body case with NON-identical nanoparticles (as some of the authors have demonstrated in their previous works), the presence of dissipation played a key role in obtaining non-reciprocity. That is, when light is injected to the two nanoparticles A and B, the light that scatters from A (B) propagates to optical force on B (A) to give rise to forces acting between the two nanoparticles. When A and B have different absorption rates, the amount of light scattering is different, giving rise to non-reciprocal interactions. Am I right that this situation (i.e. the absorption plays a crucial role for non-reciprocity to emerge) holds true for the N-body non-reciprocal interaction demonstrated in this work? If so, I believe it would be helpful to emphasize a little more explicitly (probably in the first paragraph of the “Results” section) that absorption plays a crucial role. If not, it would be interesting to add a discussion on this intriguing difference.

Response #2: We appreciate the reviewer’s insightful comment regarding the impact of the absorbing property of nanoparticle materials on non-reciprocal optical forces occurring in both two-body or N-body systems. As the reviewer commented, in a two-body system, the non-reciprocal optical forces rely on the differing polarizabilities (α_1, α_2) of the constituent nanoparticles. Based on Aristide Dogariu’s work (1), the non-reciprocal optical force is proportional to the polarizabilities, expressed as $-\alpha_1' \alpha_2'' + \alpha_2' \alpha_1''$. Here, the single and double prime symbols in the superscript denote the real and imaginary parts of the polarizabilities, respectively, with the letter featuring the absorbing property of matter. This proportionality explicitly demonstrates that a nonzero imaginary part of the polarizability (i.e., the nanoparticles’ absorbing property) is necessary for generating non-reciprocal optical forces. However, it does not imply that the absorption of constituent nanoparticles must be intense. In fact, in most cases, the absorption is minor with limited contributions.

As shown in Fig. R2, which shows the optical properties of the most commonly used nanoparticle materials (i.e., Ag and Au) in optical trapping studies, they both exhibit minimal absorption compared to their scattering strength when excited in their off-resonance spectral range. In our experiments, we chose a trapping laser wavelength of 770 nm, which is significantly distant from the dipolar resonance of a 150 nm dia. Ag nanoparticle. At this spectral position (indicated by the red vertical dashed line in Fig. R1a), the Ag nanoparticle’s absorption is nearly zero and thus negligible, indicating that its contribution to optical forces can be justifiably ignored in the N-body system proposed here.

Nevertheless, we acknowledge the reviewer’s point that a strong absorbing property of nanoparticles could play a crucial role in N-body nonreciprocal interactions if the constituents were excited resonantly; however, this aspect is not considered in our current study due to the fact that the investigated Ag nanoparticles exhibit trivial absorption at the trapping laser wavelength.

Rather, the main source of the putative non-reciprocal force in the case of 2-nanoparticle OM systems is the phase shift of the scattered waves. Since the nanoparticles scatter light elastically, the resultant interference between the nanoparticle scattering can change the direction of the resulting wavefront. The argument is similar to wavefront construction using Huygens wavelets. These ideas were presented in 2 or our previous papers:

Reactive optical matter: light-induced motility in electrodynamically asymmetric nanoscale scatterers
Y Yifat, D Coursault, CW Peterson, J Parker, Y Bao, SK Gray, SA Rice, N.F. Scherer,
Light: Science & Applications 7 (1), 105 (2018).

Controlling the dynamics and optical binding of nanoparticle homodimers with transverse phase gradients
CW Peterson, J Parker, SA Rice, NF Scherer, Nano Letters 19 (2), 897-903 (2019).

We have added the text of this reply and discussion as a new section in the Supplementary Materials

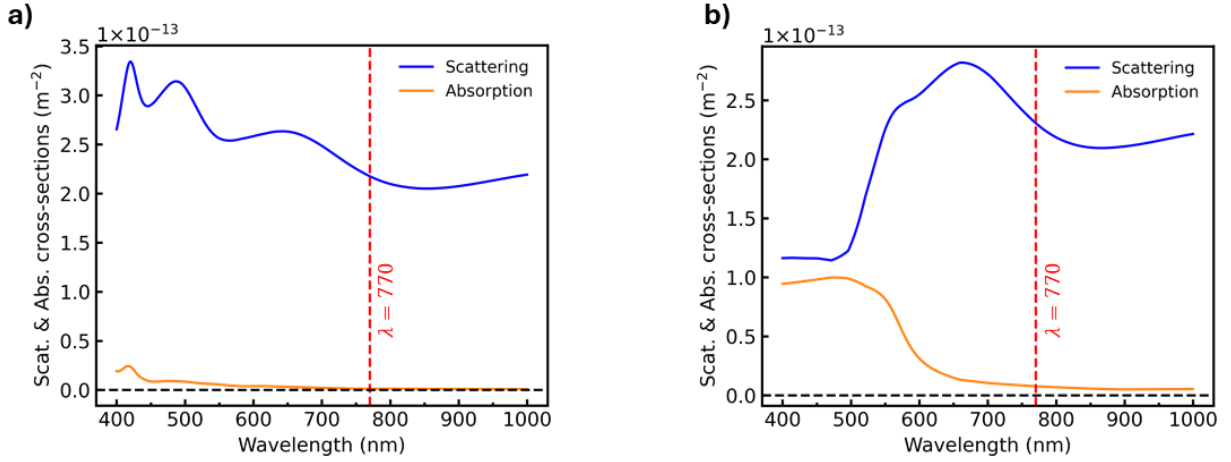


Figure R2: Absorption properties of Ag and Au nanoparticles with a diameter of 150 nm. (a) and (b) show the calculated scattering (blue curve) and absorption (orange curve) spectra of Ag and Au nanoparticles, respectively. The red and black dashed lines highlight the spectral location of the 770 nm trapping laser wavelength used in the experiment and the zero value, respectively.

Comment #3: (Slightly related to above) I was confused with the definition of α . Is α a complex number including absorption or a real number? I got confused because, in Eq. (2), the second term is proportional to α^* (with *) implying α includes an imaginary part (which physically makes sense to me), while in Fig. 2(b), α is assigned with a real number. Please clarify the definition.

Response #3: We thank the reviewer for highlighting the confusion about the polarizability notation in Fig. 2b of the main text. The nanoparticle polarizability, α , is a complex number, the strength of which is expressed in relative values with respect to the static polarizability, α_0 , for a succinct numerical representation in Fig. 2b. The static polarizability is commonly known and expressed as:

$$\alpha_0 = 4\pi a^3 \epsilon_m \epsilon_0 \frac{\epsilon(\omega) - \epsilon_m}{\epsilon(\omega) + 2\epsilon_m}, \quad (R1)$$

where a is the radius of a nanoparticle; and the $\epsilon(\omega)$, ϵ_m and ϵ_0 are the permittivities of the nanoparticle, its surrounding material, and the vacuum, respectively. To eliminate this confusion while improving the flow and readability of the manuscript for readers, we made modifications in lines 167-168 on page 7:

“For clarity, the polarizability strength is expressed as values relative to the static polarizability, α_0 [53].”

and the caption of Fig. 2:

“(b) Total azimuthal forces increase as a function of the magnitude of the single particle polarizability (relative to the static polarizability, α_0 [53]).”

Comments #4: Below Eq. (2), it would be helpful for the readers if they remarked that the second term vanishes because the particle is identical.

Response #4: We thank the reviewer’s suggestions to clarify the properties of the two-body force component defined as the first term, $|\alpha|^2/2\sum\partial_{x\xi}(\bar{\bar{G}}_{ij})$, in Eq. 2 of the main text. This force cancels out when two nanoparticles are identical in terms of their polarizabilities. As explained in lines 125-132 of the revised main text, this cancellation is rooted in the symmetrical property of the dyadic Green’s function, i.e., $\bar{\bar{G}}_{ij} = \bar{\bar{G}}_{ji}$. This symmetry ensures that the electric fields scattered by two identical nanoparticles toward each other are identical at their respective locations. Consequently, the two-body forces exerted on these two nanoparticles through the gradients of the scattered electric fields, $\partial_{x\xi}(\bar{\bar{G}}_{ij})$ and $\partial_{x\xi}(\bar{\bar{G}}_{ji})$, are equal in magnitude but opposite in direction. In contrast, the second and third terms in Eq. 2 denote 3-body forces arising from the broken parity symmetry of the configuration as shown in Fig. 1d. 3-body forces are present even among identical nanoparticles, the mechanism of which is detailed in lines 133-139 of the main text.

In response to the reviewer’s concerns, we improved the statements in lines 125-139 on page 5, which now read:

“The first term in Eq. 2 represents a strictly two-body force that cancels out when the interacting nanoparticles are identical. This cancellation occurs due to its dependences: the polarizability, α , of each particle interacting with the incident field, the gradient of the field scattered from the other constituents in the OM structure, and implicitly the induced-polarization of each nanoparticle induced by the incident and scattered fields. The dependences ensure that the pairwise (2-body) forces between two identical nanoparticles are equal in magnitude and opposite in direction as manifest in the symmetry of the dyadic Green’s function. Specifically, $\bar{\bar{G}}_{ij} = \bar{\bar{G}}_{ji}$ and $|\alpha|^2\partial_{x\xi}(\bar{\bar{G}}_{ij}) = -|\alpha|^2\partial_{x\xi}(\bar{\bar{G}}_{ji})$.

In contrast, the second and third terms in Eq. 2, which are proportional to α^3 , represent 3-body forces. Unlike 2-body forces, 3-body forces can arise even among identical nanoparticles. For instance, the term $\alpha^\bar{\bar{G}}_{ij}\alpha_{x\xi}(\bar{\bar{G}}_{ik})$ corresponds to the field scattered from particle k to the location of particle i ($\alpha_{x\xi}\bar{\bar{G}}_{ik}$) with the polarization of particle i induced by light scattered from particle j ($\alpha^*\bar{\bar{G}}_{ij}$). Simply switching indices in the second and third terms does not result in mutually canceling forces except in specific cases like a linear trimer as shown in Fig. 1(a,c), where the mirror and inversion symmetry of the configuration leads to cancellation.”*

Comment #5: The authors use the words ‘fully filled’, ‘half filled’, etc. as if there is an underlying lattice structure (such that a filled or vacant site can be defined). To my understanding, the crystalline structure is emergent due to the reciprocal piece of interaction and is not something that the authors externally imposed. It would be helpful to clarify this point in the manuscript, as the definition of ‘fully filled’ etc. was not so clear to me.

Response #5: We understand the reviewer’s confusion regarding the term “filled” since it is commonly used in solid physics to describe lattice site occupancy in crystalline materials. In our context, the “filled” describes nanoparticle configurations in the ring trap where interacting nanoparticles can fit in. As demonstrated in Fig. 2a and 2e of the main text, when Ag nanoparticles are trapped in the annular region, optical bindings between adjacent nanoparticles forms. Given the circumference of the ring trap, the interparticle separation naturally limits the maximum number of nanoparticles that can be accommodated. Once this maximum is reached, we describe the ring trap as being “fully filled”; otherwise, it is “partially

filled”. A specific scenario of the partial fill is termed “half filled”, indicating that half of the ring trap contains nanoparticles, as shown in Fig. 2c of the main text.

We added the following sentence to the text in lines 188-190 on page 8:

“By “fully filled rings” we mean the configuration of nanoparticle in the ring trap with the maximum number of electrodynamically interacting nanoparticles possible for a fixed ring radius, as demonstrated in Fig. 2a and 2e.”

Reviewer #3 (Remarks to the Author):

Comment #1: The authors present interesting results on the non-reciprocal forces in a N-body system that induces active motion of a cluster of nanoparticles confined within an annular optical beam. They show that due to the optical confinement of the Ag particles on a ring their individual scattering leads to a breaking of the symmetry between each and other and overall a net force that creates circular motion depending on the polarisation of the trapping beams.

The experimental results are sound and clearly supported by rigorous simulations. Great care has been taken to ensure that no other optical effects such as aberrations to interfere with the induction of a net force. Whether the basis of the simulations are reasonable for such experimental system is outside my scope of expertise.

Response #1: We thank the review’s thoughtful summary and positive feedback on our work. We are pleased that the reviewer found our experimental results sound and well-supported by our rigorous simulations. We also appreciate the review’s candor. Our computational modeling and simulations closely represent the experimental setup and phenomena observed.

Comment #2: However, the current version of the manuscript does not fully justify the claims of the paper on symmetry breaking of N-body systems in optical matter systems. Although the authors clearly demonstrate N-body forces within their experimental system, there are already several examples of such systems, also indicated by the references provided. A quick literature research showed up other examples of an optical system (Thermal-motion-induced non-reciprocal quantum optical system, Nonadditivity of critical Casimir forces) where symmetry is being broken and lead to the creation of additional forces in the system. A clearer justification within the context of the present literature is required to better support the new findings the authors try to claim.

In its current state, I fail to see the significant advancements of our understanding on non-reciprocal to the field of, e.g., active matter.

Response #2: We would like to first thank the reviewer for acknowledging the clarity of the N-body effect we demonstrated using the ring optical trapping configuration. However, at the same time, the reviewer raises a concern about the insufficiency of the justification for novelty of the proposed symmetry breaking within N-body interactions in a broader context of other active matter systems, particularly considering the reviewer’s two references (2, 3).

In response to this concern, we emphasize that the primary contribution of our work lies in offering a new perspective on parity-symmetry breaking that facilitates the generation of a nonreciprocal electrodynamic forces among many particles with homogeneous properties. In this regard, we found, after

carefully reviewing the two recommended references (2, 3), that they do not closely relate to the essence of our work. Specifically, the first reference (2) mainly explores three-body potentials arising from asymmetric surface boundary conditions (i.e., the inhomogeneity) of colloidal particles. The second reference (3) addresses nonreciprocity in optical transmission due to atomic thermal motion within the interacting medium.

We believe that our reported electrodynamic symmetry breaking is distinct and novel in comparison to these referenced works. The symmetry breaking discussed in our manuscript is uniquely induced by the asymmetric configuration of active matter systems, rather than the inhomogeneity of constituents as commonly explored in the literature (including the aforementioned references (2, 3)). This configuration-originated symmetry breaking and its resulting non-reciprocal forces and collective mechanical properties (dynamics) arising from N-body forces clearly advance active matter studies. This advancement has also been recognized by *Reviewer #1*, who noted that our proposed nonreciprocal electrodynamic forces, not based on asymmetric pairwise interactions, have not been previously discussed and could open new avenues for nanoparticle manipulation amongst other opportunities.

While we do not completely agree with the review's comment, we appreciate its certain aspects and have incorporated them into our revised introduction.

Comment #3: Overall, the manuscript requires significant restructuring and modification of the introduction, results and discussion section to provide better readability and enhancing the clarity of the presented results. A large part of the current introduction has been dedicated to the results of the paper while the literature has been only briefly mentioned. Here, a larger introduction into the topic of non-reciprocal forces would greatly benefit the reader to place the authors work into context. Also why symmetry and the breaking of it are important, for instance in the field of active matter, is lacking in further depth. This should allow the reader to better understand how the results could be used in different communities. A further description of the current limitations of other studies on N-body forces, and how the authors work tackles those is missing as well. In the beginning of the results section a flow from the context of the literature into the experimental system should be provided. Here a brief description of the experimental system and other parameters used would provide a basis for the results presented later. Without it, as a reader, one is quickly lost in the details of the results and requires significant forth and back between the description of figures and crucially missing info in written either in the methods section or hidden in the Supplementary Information. More clear referencing from the main manuscript to the SI is needed to allow for quick referencing. In contrast to the introduction, the discussion section recites again the literature and challenges, explaining only here the significance of their work. This should be placed instead in the introduction section. This space should be dedicated better to the discussion of the results, other aspects to be considered, such as the influence of radius of the optical beam. Detailed questions that could partly be answered here are provided below.

Response #3: We appreciate the detailed suggestions provided by the reviewer for improving the readability of our manuscript. In response to these comments and suggestions, we have made many significant revisions (please see the revised version). We believe that these revisions have significantly enhanced the quality of the manuscript and have thoroughly addressed all concerns raised in the reviews.

In lines 40-55 on page 2:

“Despite these successes that are based on 2-body interactions, our understanding of symmetry breaking-associated N-body forces remains a challenge. For example, other than the Axilrod-Teller three-body Van der Waals force [45-47], little is known of what symmetries must be broken to produce N-body forces. This limited knowledge makes it difficult to predict or quantify how symmetry breaking at the microscopic level affects macroscopic behavior, especially in nonrelativistic, non-equilibrium systems where long-range interactions (like electromagnetism) dominate. Moreover, theoretical modeling often

struggles to fully capture and elucidate N-body forces in complex systems, as the complexity of the interactions grows exponentially with the number of particles involved. Therefore, statistical approaches have been serving as the primary theoretical treatment of these systems [48].

Although some studies have explored non-reciprocal effects in various systems of asymmetric interactions [49-51], current theoretical frameworks fall short in demonstrating the dynamics resulting from N-body interactions among particles with homogeneous properties. This is largely because existing symmetry-breaking models are rooted in the inhomogeneity of interacting particles, such as distinct polarizabilities [6, 29, 31, 32] and surface boundary conditions [51]. Addressing these limitations is essential for advancing our understanding of collective behaviors across diverse fields.”

In lines 100-102 on page 4:

“Figure 1(e,f) shows that broken mirror and/or inversion symmetry is manifest in the EM field intensity scattered amongst the particles (see Sect. 2 in the Supplementary Information for more details)”

In lines 140-141 on page 6:

“In order to make and maintain the bent configuration of the trimer stable, we utilize an optical ring trap [28, 30, 31] of radius R (see Section 7 in the Supplementary Information for details).”

In lines 148-152 on page 5-6:

“Moreover, the ring configuration (optical ring trap) enables the addition of multiple particles in a curved arrangement where the increasing number of optically-induced polarizations in the nanoparticles enhances the system's scattering through electromagnetic coupling among the constituent particles. Therefore, the non-reciprocal (N-body) forces increase for progressively larger continuous (contiguous) OM structures (see Sections 3 and 5 in the Supplementary Information).”

In lines 179-181 on page 8:

“Further details on the proportionality of F_ϕ with respect to the increasing number of particles (accommodated by the expanding radius of the ring trap) is provided in Section 3 of the Supplementary Information.”

In lines 190-193 on page 8:

“In addition to the dependence of the N-body forces on the strong electromagnetic interactions as the particle number increases, the ellipticity of the polarization of incident light also influences the performance of the N-body effect. This is discussed in Section 6 of the Supplementary Information.”

In lines 201-204 on page 8:

“The fluctuations in the trajectories are due to thermal Brownian noise from the environment; i.e., the water solution. See Sections 8-16 in the Supplementary Information for more details on the transport behavior of the particles in the ring.”

In lines 224-228 on page 10:

“The quantitative differences between the velocities in experiment and simulations arise from smaller

power density in experiment ($\approx 10^8 \text{ W/m}^2$ in experiment and 10^{10} W/m^2 in simulation) and frictional drag with the nearby charged glass surface in experiments (see Sections 10, 13, and 14 in the Supplementary Information for the effect of frictional drag on the motion of the particles in experiment)."

Comment #4: The authors have used Ag nanoparticles for their study. Could they better justify the choice of such material, metallic vs. dielectric particles (commonly used in experiments), and why Ag in particular?

Response #4: We thank the reviewer for questioning the criteria we used for the material selection in the present work. As Eqs. 1 and 2 indicate in the main text, the 3-body (or N-body) forces are the cube of the polarizability of interacting nanoparticles, meaning that the N-body effect benefits from using materials that allow for large induced polarizabilities as nanoparticles. Plasmonic nanoparticles (e.g., Ag or Au, etc.) are the ideal choices. At optical frequencies, plasmonic resonances imbue metallic nanoparticles with large polarizabilities and consequently they exhibit a stronger N-body effect as compared to their dielectric counterparts. We have shown this in Fig. R1 in Response #3 to the Comment #3 of Reviewer #1: the 3-body force arising from/acting on Ag nanoparticles is nearly one order magnitude larger than that for Si nanoparticles when they have the same diameters and for the same optical excitation condition.

Of course, other metallic nanoparticles can be suitable candidates for the study; for instance Au, which is commonly used in the study of optical matter. It is noteworthy that Ag nanoparticle relative to the Au ones exhibit much less absorption (see Fig. R2 in the Response #2 to the Comment #2 of Reviewer #2), which dramatically prevent optical-induced thermo-phoretic effects.

This was the main reason why we conducted our study with Ag nanoparticles. While the N-body broken symmetry force studied here will arise in various nanoparticle materials, selecting Ag nanoparticles for investigating the N-body force is because of its strong optical polarizability and negligible absorbance.

Comment #5: What other forces could be at play in such systems that could drive particle motion besides optical gradients? What about thermal forces due to absorption of light on nanoparticles? For example: <https://www.nature.com/articles/s41467-022-28212-z>

Response #5: We appreciate the reviewer's concern regarding hydrodynamic forces arising from optically induced thermos-osmotic flows, which, the reviewer believes, potentially influenced by the absorbing properties of metallic nanoparticles in our optical matter system. Indeed, the study the reviewer referenced (4) demonstrates that optically induced temperature gradients near a 50 nm thin Au film, illuminated by a 532 nm laser beam, significantly influence particle manipulation. This effect is primarily due to the plasmonic resonance of the Au film closely aligning with the excitation wavelength, resulting in strong absorption at this wavelength, as explained by the Kramers-Kronig relations (5, 6). Such on-resonance excitation generates local heating on the Au film, subsequently causing convective flows at the Au-water interfaces, which impact particle dynamics.

However, in the conditions of our optical tweezer studies, such optically induced thermal effects can be safely disregarded for two reasons. Firstly, as discussed in the previous response (*Response #4*) and demonstrated in Fig. R2 in *Response #2* to *Comment #2* of *Reviewer #2*, the investigated Ag nanoparticles, when excited off-resonance, exhibit negligible absorption and very small absorbance near-resonance, which is the condition of our experiment. Secondly, the glass-water interface, proximate to where the ring-configured optical matter forms, does not absorb incident light nor generate local heat. It is totally transparent at the laser wavelength used in our study. Finally, from our extensive experience with studies of Ag nanoparticles in similar experiments, we have never detected any enhanced or directed motion that might be associated with thermal gradients. Therefore, we believe that we have excluded any forces associated with optically induced thermal effects in our analysis.

Comment #6: From Figure 1h, it is not clear to me why there is only a force component along x here and not y as well. Shouldn't the force be along azimuthal as presented later on as well?

Response #6: We appreciate the reviewer's insightful question regarding the y-component of the 3-body force. As shown in Figs. 1c and 1d of the main text, when the linear trimer is bent at a small angle (e.g., less than 10° , consistent with the actual conditions of our experiments), mirror (parity) symmetry along the x-axis is broken, while it remains along the y-axis. This broken symmetry results in the net (i.e., 3-body) force being predominantly oriented along the x-axis. As shown in Fig. R3, at these small bending angles, the y-component of the 3-body force is nearly zero. Therefore, in our ring optical trap, the y-components of the N-body forces, which are radial, are significantly overshadowed by the stronger, radially directed dipole (intensity gradient) forces. This justifies our focus primarily on the x-component (i.e., the azimuthal direction) of the N-body forces in the analysis. Furthermore, in the experiments the nanoparticles are confined to the optical ring trap and cannot move appreciably in the radial direction.

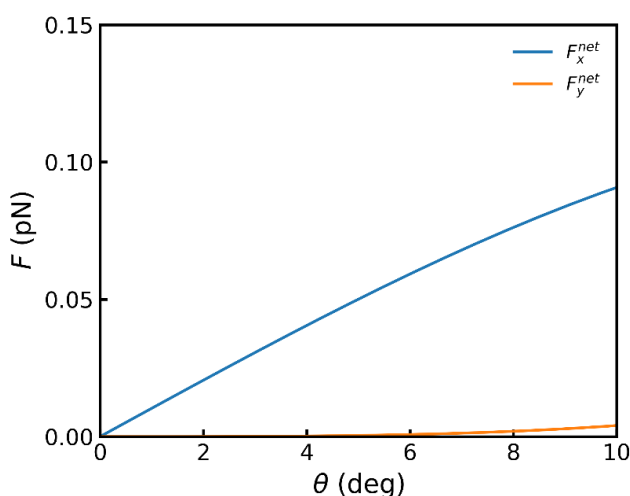


Figure R3. Three-body optical forces on a trimer composed of 150 dia. Ag nanoparticles along x- and y-axis with bending angles ranging from 0° to 10° . This range of bending angles represent the actual bending angles observed in the experiments of ring-configured optical matter structures.

Comment #7: Have the authors analysed the trapping stiffness of the optical trap along the radial coordinate?

Response #7: The trapping stiffness along the radial coordinate can be inferred from the Fig. S16a of the Supplementary Information and is added in the Section 16 of the Supplementary Information:

“Additionally, the radial position distributions shown in Fig.S16(a) allow determines the stiffness coefficients, k_r , of the experimental optical trap in the radial direction. The radial stiffness coefficients are determined to be approximately $1.78 \text{ pN}/\mu\text{m}$, $1.79 \text{ pN}/\mu\text{m}$, $1.81 \text{ pN}/\mu\text{m}$ for the cases of 44 particles, 43 particles, and 43 particles with gap, respectively.”

Comment #8: Line 94: What does quickly restored refer to here? Can they provide a number?

Response #8: The term "quickly restored" is a qualitative description of how a trimer dynamically responds to a physical bend as a result of its electromagnetic interactions. This dynamic response can vary along with random changes (or fluctuations) in various environmental factors, which, however, do not alter the

fundamental properties of the N-body effect demonstrated here. Therefore, providing a precise numerical to define the "quickly restored" would not offer additional useful information about the strengths or performance of the N-body effect.

After careful consideration of the reviewer's suggestion, we have decided to omit the word "quickly" from the text and simply say "restored" to avoid confusion.

Comment #8: The authors provide only experimental data for filled rings of two radii and it three configurations. To support their claim and simulations it would be beneficially to also show experimental data of lower number of particles N.

Response #8: We thank the reviewer for the suggestion to include more experimental data to support our findings regarding the N-body effect with a lower number of nanoparticles. In this manuscript, we have provided data from three representative ring radii and particle numbers in our experiments: $R=4.1\ \mu\text{m}$, $N=44$ (fully filled); $R=4.1\ \mu\text{m}$, $N=43$ (partially filled); and $R=6.3\ \mu\text{m}$, $N=65$ (fully filled), as illustrated in Sections. 12 and 13 in the Supplementary Information. We observed that the consistent manifestation of the N-body effect requires optical matter structures to remain stable, regardless of whether the rings are partially or fully filled.

In scenarios where the optical ring trap contains a smaller number of nanoparticles (i.e., is partially filled), thermal fluctuations can easily disrupt the optical binding among the constituent nanoparticles in the azimuthal direction, intermittently impeding the N-body dynamics. Conversely, a fully filled ring overcomes these thermal-induced structural fluctuations by providing azimuthal bounding, thereby ensuring the stability of the optical bindings and hence the integrity of the optical matter structures.

Furthermore, the N-body force scales with the number of particles so we empirically found that the effect is most readily observable for larger rings with more particle constituents. Also, other effects due to deviations from the paraxial approximation can cause deviations from the theoretical description that we developed.

Given these reasons and our complete analysis, we believe that our current experimental data set sufficiently supports our claims regarding the N-body effect occurring under various ring radii and filling conditions. After careful consideration, we have decided not to include additional experimental data in order to maintain focus and clarity within our presented findings.

Comment #9: Figure 2c: What is the reason for the bent in the graph just above 20 particles? What is the dependence on the distance between particles here for cases where particles are not fully filling the ring?

Response #9: This is an interesting question that we have also wondered about. The capacity of a ring optical trap is determined by the number of optical binding interactions that can be accommodated within its circumference. Optical binding is a laser-induced inter-particle separation, typically about one wavelength of light in the medium. We believe that the non-linear trend in the N-body force vs. particle number results from changes in inter-particle separations when the rings are partially filled. As we show in Figure 3, the angular velocity of the nearly-filled and filled rings are significantly different and have found that this is associated with the inter-particle separation deviating from an ideal condition that begets maximum constructive interference. There could be other statistical effects, such as the combinatorial of the number of N-body combinations saturating.

In our electrodynamics-Langevin dynamics (EDLD) simulations, larger diameter ring traps can accommodate more particles, but require significantly more computational resources. Increasing the number of particles in the simulation does not necessarily provide additional insights into N-body dynamics or

enhance the interpretation of our experimental findings; rather, it incurs unnecessary computational costs. The ring trap setup shown in Fig. 2c of the main text, accommodating up to 28 nanoparticles, offers computational efficiency and provides sufficient information on the dynamics of the ring-configured optical matter structure.

Finally, in both experiment and EDLD simulations, when the ring optical trap is partially filled, thermal fluctuations may disrupt the optical binding, resulting in random larger distances where optical binding does not occur.

Comment #10: Figure 2d: Why are the distributions of the different sizes here?

Response #10: We thank the reviewer's query about the distributions of the velocities (PDF) for different trapping conditions shown in Fig. 2d of the main text. The azimuthal driving (i.e., N-body) force varies along with the fluctuations in the positions and spacing between particles (7, 8), with thermal fluctuations consequently producing a distribution of total azimuthal driving forces. The variance in these distributions decreases as the number of particles increases. This reduction is because optical binding forces reinforce one another in regularly spaced chains of particles, and longer chains tend to be trapped more rigidly (9, 10).

Comment #11: What was the reasoning behind not using the same power density for the simulations in order to make the comparison with experiments?

Response #11: We appreciate the reviewer's question regarding our not matching the exact power density in our EDLD simulations with that used in the experiments. Ideally, replicating all experimental parameters would be preferable. However, we were not striving for quantitative comparisons since the experiments have a frictional interaction with the nearby glass coverslip that is not practical to simulate (other than to guess what it is). The primary goal of our EDLD simulations, as shown in Figs. 3f and 3g of the main text, is to capture and interpret the essential dynamic properties of the ring-configured optical matter structures (illustrated in Figs. 3d and 3e), modeled in our Generalized Multiparticle Mie Theory (GMMT)-Langevin dynamics model. Therefore, for reduced computational cost we conducted simulations with somewhat larger power densities and concomitantly less computer simulation time.

We discovered that varying power densities within a reasonable range did not lead to simulation conclusions that contradicted our experimental observations. Moreover, setting the exact same power density as in the experiments would require precise determination of other relevant parameters, including experimental substrate friction coefficient, solution damping coefficient, stochastic force, temperature, and the timescale of Brownian motions, which are not always feasible to ascertain with absolute accuracy.

Given the strong agreement between the simulation results and experimental outcomes, and the simulations' relative insensitivity to changes in power density, we believe that our simulation settings effectively isolated the key mechanisms driving the observed phenomena. We acknowledge that further investigation into the precise effects of laser power density could be a valuable focus for future research.

Comment #12: SI 3: The statement for the step-like behaviour in Fig. S2 would benefit from a more clear illustration of when particles had to be added depending on the ratio between ring and particle diameter.

Response #12: We appreciate the suggestion to enhance the explanation of the step-like behavior of the azimuthal forces shown in Fig. S2 of the Supplementary Information. We would like to clarify that although the plot shows the force as a function of radius, it actually also reflects the number of particles, N . As N increases, the radius also increases (resulting in a decrease in bend angle) to accommodate this change. Consequently, both the interparticle separation and ring radius adjust to optimize chord length (optical binding) for N-body interactions. The interparticle separation changes in a stepwise manner, mirroring the

trend observed in the force. We have updated the statement accordingly in Section 3 of the Supplementary Information:

“As the radius of the ring is increased the number of particles that can be accommodated along the expanding circumference of the ring increases discretely. Additionally, the interparticle separation, which also affects the N-body force $F_\phi(R, N)$ as demonstrated in Eq. 1 of the main text, varies periodically due to alternating compression and relaxation (dilation) of interparticle separation as the radius, R , and circumference are increased and also the particle number, N . Discrete jumps in the force are related to obtaining more ideal interparticle separations for certain ratios of ring trap circumference to the number of nanoparticle constituents. The upward trend in $F_\phi(R, N)$ is an additive effect of increasing particle number.”

Minor comments:

Comment #13: Line 41: Here N-body ED forces are immediately assumed which is not obvious to the reader. A better clarification is required.

Response #13: The statement has been revised for better clarity and is now read in lines 56-61 on page 2-3:

“In this paper we consider optical matter (OM) systems [5, 7, 21-29] of three or more identical Ag nanoparticles in a laser trap with optical forces arising from N-body electrodynamic interactions among them. We show that breaking the spatial symmetries of 3 or more nanoparticles gives rise to non-reciprocal force that, in turn, determines the collective behavior of the system. The electrodynamic interactions and N-body forces are calculated using rigorous Generalized Multiparticle Mie Theory (GMMT) simulations and Maxwell stress tensor analysis.”

Comment #14: Line 50: Not clear what bent configuration refers to here.

Response #14: The bent configuration refers to the one shown in Fig. 1d. For a better understanding, the statement has been revised and now reads in lines 63-69 on page 3:

“The N-body non-reciprocal forces only arise for three or more particles maintained in a bent configuration (see Fig. 1d) where the OM system has broken mirror (reflection) symmetry. The requisite symmetry breaking is realized in experiment by trapping many Ag nanoparticles using an optical ring trap to sustain the bent configuration [30]. Having assured the absence of transverse phase gradients in the ring that would drive the nanoparticles [29, 31, 32], the observed collective azimuthal motion of the ring of optically bound nanoparticles is due to these N-body non-reciprocal forces.”

Comment #15: Line 65: Linear multimers are not shown here, so why mention it?

Response #15: We want to emphasize that the phenomenon applies not only to linear trimers but also to linear chains of one-dimensional optical matter structures, as reported in our previous work (10). For clarification, the revised statement in lines 81-83 on page 3 now reads:

“Figure 1(c,d) shows the same for an incident plane wave that is right-handed circularly (RHC) polarized. In the case of a linear trimer (or linear multimer [52]) (Fig. 1(a)), the induced polarization between the particles has mirror symmetry along both the x and y directions.”

Comment #16: Line 73: Justification for that statement missing.

Response #16: The statement (in lines 91-96 on page 3-4) has been revised for clearer understanding and

now reads:

“By contrast, in the case of right-handed circularly (RHC) polarized light, as shown in Fig. 1(c,d), the induced dipoles (i.e., induced-polarizations) are elliptical with the same handedness as the incident light. The elliptical (non-circular) shape of the induced polarization results from the phase delay associated with light scattered between the particles. The induced dipoles would be circular (i.e., have equal magnitudes in all orientations) in the case of non-interacting particles illuminated by circularly polarized light, and there would be no net force other than single particle rotation.”

Comment #17: Figure 1d/S1: Why are the orientations of the elliptical polarisations not the same? Is that on purpose or an illustration error?

Response #17: We thank the reviewer’s attention to the elliptical shape of the induced polarization of the trimer shown in Fig1 of the main text. As demonstrated, the ellipticity of the polarization of an induced dipole results directly from the scattering of light between the particles. This relation is manifested in Eq. 4 in the Methods section. When the timer is aligned in a row, the second term of Eq. 4, which represents pairwise (two-body) interactions dominates and preserves reciprocity (and thereby the inversion symmetry) in their mutual scattering. This explains the identical and aligned ellipticities in the trimer’s induced polarizations. As the linear configuration is disrupted by bending, the third term of Eq. 4, which defines three-body interactions, starts to influence the system. The impact of this term is linearly proportional to the bending angle for small values, as evidenced by the empirical formula displayed in Fig. 1g. However, unlike the two-body interaction term, the three-body interaction term breaks the inversion symmetry of the system, resulting in nonreciprocal scattering among the particles. This nonreciprocal scattering accounts for the differing orientations of the induced dipoles in the bent configuration of the trimer shown in Fig 1d. Note that a related effect is shown in the phase of the fields around the nanoparticles.

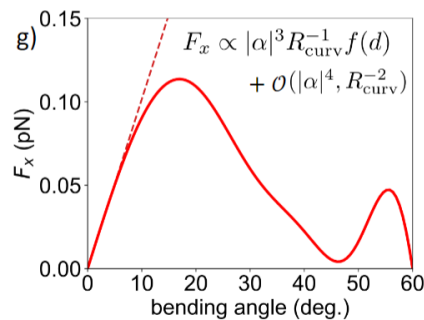
Comment #18: Figure 1g: Not clear to everyone what h.o. stands for, better to use same annotation as in Eqn 2 throughout whole paper.

Response #18: The term “h.o.” of Eq. 1 stands for high order. In response to the reviewer’s suggestion, we replaced it with the notation \mathcal{O} to ensure consistency throughout the manuscript. The revised equation and figure now show:

Equation 1 expresses now:

$$F_x \propto |\alpha|^3 R_{\text{curv}}^{-1} f(d) + \mathcal{O}(|\alpha|^4, R_{\text{curv}}^{-2})$$

Figure 1g is now:



Comment #19: Figure 1e,f scale bars missing.

Response #19: The spatial scale of the representation of the trimers remains consistent throughout Figs. 1a-1f, with a focus on qualitatively describing the linear and bent trimer configurations, rather than reflecting the actual size ratio of nanoparticles vs. inter-particle separation. Adding a scale bar to Figs. 1e and 1f could create confusion for readers when interpreting the size ratios shown in Figs. 1a-1d. Moreover, the absence of a scale bar does not hinder the readers' ability to understand the preservation or breaking of inversion symmetry in the electromagnetic fields for the linear and bent trimer configurations. Therefore, we decided to not include scale bar in Figs. 1e and 1f.

Comment #20: Line 97: Not clear what x_ξ stands for.

Response #20: x_ξ is used to denote the direction along which the gradient (partial derivative) of the Green's function $\bar{\bar{G}}_{ij}$, indicating the gradient of electromagnetic field scattered from adjacent particle j at the position of particle i .

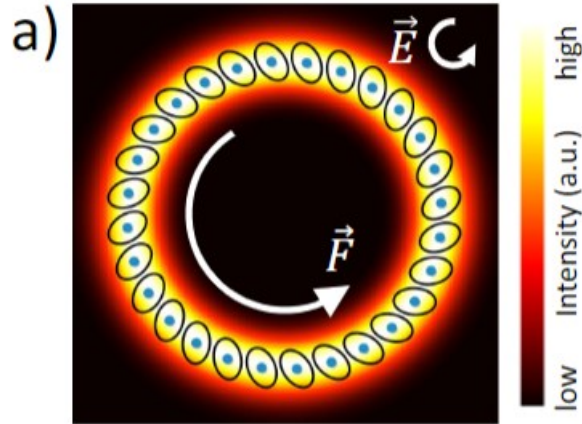
Comment #21: Line 123: spacing error

Response #21: Now, it reads in line 156-158 on page 6:

“Furthermore, the N -body interactions in the ring cause a N -body induced-polarization around the ring that behaves as a unidirectionally propagating wave in the clockwise direction resulting in a broken spatial symmetry [9, 19].”

Comment #22: Figure 2a: Missing color bar

Response #22: We thank the reviewer for this. The color bar has been added to Fig 2a, as shown below:



Comment #23: Figure 2b: F_ϕ should be noted here as variable as throughout the rest of the manuscript. Why the choice of 70x? A better way of presenting the data should be chosen without arbitrary scalings such as normalisations of the curves.

Response #23: Thank you for the suggestion. Using the term "azimuthal force" would indeed be more straightforward for readers compared to " F_ϕ ." Multiplying the 3-body electromagnetic force by 70X allows for a direct comparison with the N -body forces in the ring-configured optical matter structure. Additionally,

this multiplication provides a clear sense of how small the 3-body force is compared to the N-body force, which would not be as apparent if normalization were used.

Comment #24: Figure 2 caption: Weird notation of diameter, better way $d=150\text{nm}$, Not consistent notation of time intervals between figure and caption.

Response #24: We appreciate the reviewer's concern regarding the notation of diameter. We have verified that this notation is correct and have chosen to retain it to maintain consistency with our previous publications. Regarding the inconsistency in the time intervals shown in Figs. 2e and 2f and their captions, we have rounded the numbers to two decimal places. This approximation should not lead to any misunderstandings.

Comment #25: Fig. S9: Would be good to see some error bars as individual trajectories should vary quite significantly as indicated by large distributions in the velocities before.

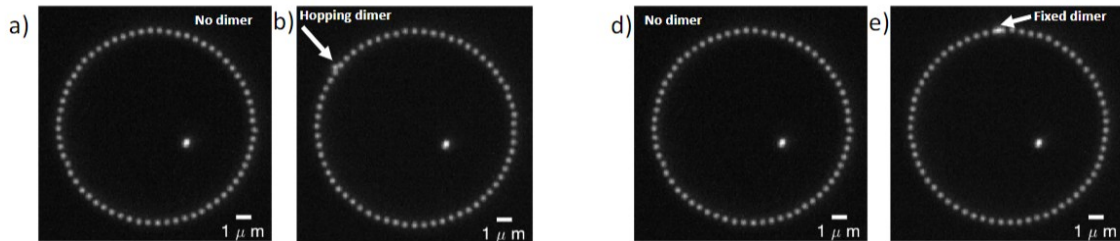
Response #25: We appreciate the reviewer's thoughtful suggestion to add error bars to the mean-squared displacement (MSD) plot, considering the bandwidth of the velocity distribution of the nanoparticles. After careful consideration, we believe that adding error bars in this case (as well as in many other MSD plots within the Supplement Information) may not provide additional useful insights. Instead, it could potentially distract from the primary purpose of the plots per their demonstrative nature.

In Fig. S9 of the Supplementary Information, the focus is not to illustrate the driven transport behavior of nanoparticles coupled electromagnetically within the ring trap. Rather, the main objective of these two plots is to compare two MSD calculation methods that are more effective in accounting for the rapid thermal motion of individual nanoparticles at short timescale. The core aspects of this comparison are well-captured in the current presentation.

While we are grateful for the reviewer's constructive feedback, we elect to maintain the plots in their current form since we believe they best serve the intended purpose of the data presentation.

Comment #26: Fig. S12 scale bars not clearly visible.

Response #26: We thank the reviewer for the comment. The scale bars have been carefully adjusted in Fig S12(a-e) to improve their visibility, as shown below:



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