Reply to "Comment on 'Unified treatment of nonlinear optical force in laser trapping of dielectric particles of varying sizes'"

Anita Devi[1,*] and Arijit K. De[2,†]

[1]Condensed Phase Dynamics Group, Department of Physical Sciences, Indian Institute of Science Education and Research (IISER) Mohali, Knowledge City, Sector 81, SAS Nagar, Punjab 140306, India

[2]Condensed Phase Dynamics Group, Department of Chemical Sciences, Indian Institute of Science Education and Research (IISER) Mohali, Knowledge City, Sector 81, SAS Nagar, Punjab 140306, India

(Received 9 June 2022; accepted 16 June 2022; published 11 July 2022)

In this Reply, we set out a discussion on the strength as well as the plausible shortcomings of *generalized Lorenz-Mie theory* in estimating the trapping force on a particle with an emphasis on the implications of ultrashort pulsed excitation.

DOI: 10.1103/PhysRevResearch.4.038002

We thank the authors for the timely Comment [1]. As mentioned in the Comment, we would like to reiterate that the choice of methods for numerically evaluating the beam-shaping coefficients (BSCs) in generalized Lorenz-Mie theory (GLMT) may produce results, which using localized approximations [2–4], were shown in Reference [5] to quantitatively differ from that obtained by exact Mie theory (EMT) [6], specifically for larger micron-sized particles or for any particle under ultrashort pulsed excitation. In particular, warnings for the use of localized approximations have been recently published for beams exhibiting axicon angles and/or topological charges [7–10], although there are strong evidences of the efficiency of localized approximations in other circumstances (e.g., Refs. [11,12] and references therein). Thus, our inference drawn on the possible limitations of localized approximations is still correct which, in no way, means that GLMT is not a rigorous theory as aptly pointed out by the authors [1].

On the other hand, despite the possible limitations, the localized approximations turned out to be quite useful to qualitatively perceive novel phenomena, such as "trap splitting" (facilitating trapping of multiple particles) or "Fano resonance" (responsible for negative optical scattering force) especially under ultrashort pulsed excitation [13–16] which, using dipole approximation [17], were not captured [18–19]. Obviously, either GLMT (using methods other than localized approximations for evaluating BSCs) or EMT is expected to produce similar quantitative results which are more accurate than those obtained from GLMT and need further investigation. However, it is our observation that, although exact in formulation, neither GLMT nor EMT, fully incorporates specific effects (for example, force propagation along the surface) for which different theoretical formalisms (for example, extended boundary condition method [20–22]) are to be followed.

It is to be noted that whereas comparing with experimental findings, even EMT showed qualitative agreement only and the quantitative disagreement was ascribed to specific hydrodynamic effects, for example, laser-induced heating leading to convection/thermophoretic effects [23,24]; photophoretic effects may also contribute to this discrepancy.

Finally, we would like to emphasize that, although there have been previous studies on scattering of ultrashort laser pulses by particles [25–28], none of these included the ensuing optical and thermal nonlinear force/potential on the particles which has been the focus of our research [5,13–16,18,19,23,24,29–36].

We sincerely thank G. Gouesbet for bringing their attention to the point discussed in the Comment. During preparation of this response, he was contacted. It was indeed a pleasure to have the numerous stimulating conversations over emails.

*Present address: Department of Physics, University of Alberta, 85 Avenue, Edmonton, Alberta T6G 2R3, Canada; adevi@ualberta.ca

†akde@iisermohali.ac.in

[1] G. Gouesbet, Comments on "Unified treatment of nonlinear optical force in laser trapping of dielectric particles of varying sizes", Phys. Rev. Research **4**, 038001 (2022).
[2] G. Gouesbet, G. Grehan, and B. Maheu, Computations of the gn coefficients in the generalized lorenz-mie theory using three different methods, Appl. Opt. **27**, 4874 (1988).
[3] G. Gouesbet, G. Grehan, and B. Maheu, Localized interpretation to compute all the coefficients gnm in the generalized lorenz–mie theory, J. Opt. Soc. Am. A **7**, 998 (1990).
[4] G. Gouesbet and G. Gréhan, *Generalized Lorenz-Mie Theories* (Springer, Berlin, 2011), Vol. 31.
[5] A. Devi and A. K. De, Unified treatment of nonlinear optical force in laser trapping of dielectric particles of varying sizes, Phys. Rev. Research **3**, 033074 (2021).
[6] P. M. Neto and H. M. Nussenzveig, Theory of optical tweezers, Europhys. Lett. **50**, 702 (2000).
[7] G. Gouesbet, On the validity of localized approximations for bessel beams: All n-bessel beams are identically equal to zero, J. Quant. Spectrosc. Radiat. Transfer **176**, 82 (2016).
[8] G. Gouesbet, J. A. Lock, L. A. Ambrosio, and J. J. Wang, On the validity of localized approximation for an on-axis zeroth-order bessel beam, J. Quant. Spectrosc. Radiat. Transfer **195**, 18 (2017).
[9] G. Gouesbet and L. A. Ambrosio, On the validity of the use of a localized approximation for helical beams I. Formal aspects, J. Quant. Spectrosc. Radiat. Transfer **208**, 12 (2018).
[10] L. A. Ambrosio and G. Gouesbet, On the validity of the use of a localized approximation for helical beams. II. Numerical aspects, J. Quant. Spectrosc. Radiat. Transfer **215**, 41 (2018).
[11] G. Gouesbet, Generalized lorenz-mie theories and mechanical effects of laser light, on the occasion of arthur ashkin's receipt of the 2018 nobel prize in physics for his pioneering work in optical levitation and manipulation: A review, J. Quant. Spectrosc. Radiat. Transfer **225**, 258 (2019).
[12] J. Shen, Y. Wang, H. Yu, L. A. Ambrosio, and G. Gouesbet, Angular spectrum representation of the bessel-gauss beam and its approximation: A comparison with the localized approximation, J. Quant. Spectrosc. Radiat. Transfer **284**, 108167 (2022).
[13] A. Devi and A. K. De, Theoretical estimation of nonlinear optical force on dielectric spherical particles of arbitrary size under femtosecond pulsed excitation, Phys. Rev. A **96**, 023856 (2017).
[14] A. Devi and A. K. De, Generalized lorenz-mie theory for the reversal of optical force in a nonlinear laser trap, Phys. Rev. A **102**, 023509 (2020).
[15] S. Yadav, A. Devi, and A. K. De, Synergistic effect of fano resonance and optical nonlinearity in laser trapping of silver nanoparticles, Phys. Rev. A **102**, 043511 (2020).
[16] A. Devi, B. Sikdar, and A. K. De, Revisiting nonlinear optical trapping of single nanoparticles using generalized lorentz-mie theory, Phys. Rev. A **105**, 053529 (2022).
[17] Y. Harada and T. Asakura, Radiation forces on a dielectric sphere in the Rayleigh scattering regime, Opt. Commun. **124**, 529 (1996).
[18] A. Devi and A. K. De, Theoretical investigation on nonlinear optical effects in laser trapping of dielectric nanoparticles with ultrafast pulsed excitation, Opt. Express **24**, 21485 (2016).
[19] A. Devi and A. K. De, Generalized description of the nonlinear optical force in laser trapping of dielectric nanoparticles, Phys. Rev. Research **2** 043378 **(4)**,(2020).
[20] T. A. Nieminen, V. L. Loke, A. B. Stilgoe, N. R. Heckenberg, and H. Rubinsztein-Dunlop, T-matrix method for modelling optical tweezers, J. Mod. Opt. **58**, 528 (2011).
[21] T. A. Nieminen, V. L. Loke, A. B. Stilgoe, G. Knöner, A. M. Brańczyk, N. R. Heckenberg, and H. Rubinsztein-Dunlop, and Optical tweezers computational toolbox, J. Opt. A: Pure Appl. Opt. **9**, S196 (2007).
[22] G. Gouesbet, T-matrix methods for electromagnetic structured beams: A commented reference database for the period 2014–2018, J. Quant. Spectrosc. Radiat. Transfer **230**, 247 (2019).
[23] A. Devi, S. Yadav, and A. K. De, Dynamics of a dielectric microsphere inside a nonlinear laser trap, Appl. Phys. Lett. **117**, 161102 (2020).
[24] A. Devi, S. Yadav, and A. K. De, Deciphering single-and multi-particle trapping dynamics under femtosecond pulsed excitation with simultaneous spatial and temporal resolution, Sci. Rep. **12**, 5373 (2022).
[25] G. Gouesbet and G. Gréhan, Generic formulation of a generalized lorenz-mie theory for a particle illuminated by laser pulses, Part. Part. Syst. Charact. **17**, 213 (2000).
[26] L. Mees, G. Gréhan, and G. Gouesbet, Time-resolved scattering diagrams for a sphere illuminated by plane wave and focused short pulses, Opt. Commun. **194**, 59 (2001).
[27] L. Mees, G. Gouesbet, and G. Gréhan, Scattering of laser pulses (plane wave and focused gaussian beam) by spheres, Appl. Opt. **40**, 2546 (2001).
[28] Y. P. Han, L. Méès, K. F. Ren, G. Gréhan, Z. S. Wu, and G. Gouesbet, Far scattered field from a spheroid under a femtosecond pulsed illumination in a generalized lorenz–mie theory framework, Opt. Commun. **231**, 71 (2004).
[29] A. Devi and A. K. De, Theoretical investigation on optical kerr effect in femtosecond laser trapping of dielectric microspheres, J. Opt. **19**, 065504 (2017).
[30] A. Devi and A. K. De, An alternate analytic formulation of optical force on a dielectric sphere in the ray optics limit, J. Opt. Soc. Am. B **35**, 244 (2018).
[31] A. Devi and A. K. De, Harnessing optical nonlinearity to control reversal of trapping force: A theoretical investigation, J. Opt. **21**, 065502 (2019).
[32] A. Devi, S. S. Nair, and A. K. De, Disappearance and reappearance of an optical trap for silver nanoparticles under femtosecond pulsed excitation: A theoretical investigation, Europhys. Lett. **126**, 28002 (2019).
[33] A. Devi and A. K. De, Simultaneous detection of two-photon fluorescence and back-scattered signal of optical trapping of dielectric nanoparticles under femtosecond pulsed excitation, J. Nanophotonics **13**, 020501 (2019).
[34] A. Devi and A. K. De, A table-top compact multimodal nonlinear laser tweezer, Opt. Commun. **482**, 126440 (2021).
[35] A. Devi, S. S. Nair, S. Yadav, and A. K. De, Controlling optical trapping of metal–dielectric hybrid nanoparticles under ultrafast pulsed excitation: A theoretical investigation, Nanoscale Adv. **3**, 3288 (2021).
[36] S. Yadav, A. Devi, and A. K. De, Enhanced optical force on multilayered dielectric nanoparticles by tuning material properties and nature of excitation: A theoretical investigation, Nanoscale Adv., doi: 10.1039/D2NA00280A.